



FACILITY FORM 802

N 67 11860	
(ACCESSION NUMBER)	(THRU)
164	1
(PAGES)	(CODE)
CR-79930	12
(NASA CR OR TMX OR AD NUMBER)	(CATEGORY)

GPO PRICE \$ \_\_\_\_\_

CFSTI PRICE(S) \$ \_\_\_\_\_

Hard copy (HC) 5.00

Microfiche (MF) 1.00

ff 653 July 85

Department of Chemical Engineering  
University of Maryland  
College Park, Maryland

FIRST ANNUAL REPORT  
ON PROGRAM TO  
COMPILE AND EVALUATE  
DIFFUSIVITY AND VISCOSITY DATA

Department of Chemical Engineering  
and  
Institute for Molecular Physics

University of Maryland  
College Park, Maryland

November, 1966

NASA Contract No. NGR-20-002-053

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## I Introduction

During the past year an interdisciplinary program was begun for the compilation and critical evaluation and of certain transport data. The project deals primarily with diffusivity and viscosity in gases, liquids and solids. The program activity has been resolved into five general areas pertaining to staff interest and background. The program of the Center is an interdisciplinary effort under the primary direction of the Institute for Molecular Physics and the Chemical Engineering Department. Dr. E. A. Mason (Molecular Physics) is Technical Director and Dr. R. B. Beckmann (Engineering College) is Executive Director.

The Chemical Engineering Department offers graduate study in materials, nuclear and chemical engineering and the Molecular Physics Institute also crosses the usual departmental lines, having staff and graduate students with physics, chemistry and engineering backgrounds. Thus the program is quite interdisciplinary and brings together a staff with a wide variety of experience and interest.

Each of the five areas is under the direction of a senior member of the faculty, with the assistance of one or more staff members. These are indicated in the following discussion. In addition each of the staff members are assisted by one or more graduate students.

The latest methods of information storage and retrieval are being employed in compiling and evaluating the data and reporting the results. In this regard close liaison with the Thermophysical Properties Center at Purdue is being maintained. In March, Dr. Marchello attended a one week workshop at Purdue and in August he attended the Gordon Research Conference on the latest methods for information storage and retrieval.

Several methods presented at the workshop and conference are being used in the project. Members of the Engineering and Science Library staff are also available to consult in this area. Similarly the University's computer facilities (IBM 360 system, IBM 7094 and IBM 1620) are available to the project.

The primary purpose of the program is to compile and evaluate diffusivity and viscosity data in gas, liquid, solid and turbulent fluid systems. The program is intended to complement the compilations of transport data by the Thermophysical Properties Research Center at Purdue University. Where it is possible, use is made of TPRC's existing and continuing literature compilation effort. This has materially reduced the literature searching chore and to provides correspondingly more time for data analysis and interpretation.

Consideration is restricted (except in the case of polymers and turbulence) to those physicochemical systems that can be characterized without ambiguity and which can provide the basis for testing existing theories and the development of new models. In short, the systems investigated are those which might appropriately be considered as Standard Reference Data. An additional objective is to bring to the attention of pure and applied scientists those areas where more work is required in order to resolve conflicting views or fill practical needs.

## II Summary

As indicated in the following sections covering the specific areas, several of the projects are fairly well along while others are just beginning. Of particular interest in this report are: the bibliography on binary gas diffusivities and the evaluated data on the He - Ar system; the bibliography on diffusivities in liquids and polymers; the data evaluation on inorganic solids; and, the turbulent transport summary.

During 1967 it is expected that a number of areas will reach the final report stage. The review of binary diffusion in gases will be completed as a NSRD Report. Compilation and evaluation of several liquid systems will be completed and available for publication. A report on the diffusivities of gases through polystyrene and polyethylene will be completed and ready for publication as will the report and diffusivities in alkali and silver halides.

A NSRD Report entitled "Review of Diffusion in Liquids, Part I" is planned for 1968 along with a NASA Report "Review of Turbulent Transport Phenomena". These will be followed in 1969 by: "Review of Diffusion in Polymers" expected to be a NASA Report; "Review of Diffusion in Inorganic Crystalline Solids" to be NSRD Report; "Review of Diffusion in Liquids, Part II"; and, "Supplemental Report on Diffusion in Gases" to be a NSRD Report.

### III BINARY DIFFUSION IN GASES

#### Work Completed

Upon completion of the most extensively studied binary dilute gas system, (see first progress report) namely He - Ar, the diffusion coefficients of gas pairs with less exhaustive data were considered. These systems were He - N<sub>2</sub>, Ar - N<sub>2</sub> and Ne - Ar, chosen partly because molecular beam scattering results were available. Diffusion measurements were correlated as a function of temperature. Results were weighted according to method.

In addition to data evaluation, bibliographies were prepared (see appendix) the first survey includes only data to-date, on noble gas pairs. The second bibliography, according to first author, is a general listing of available citations. The general bibliography also specifies the gas-pairs studied (see appendix).

#### He - N<sub>2</sub>

For the He - N<sub>2</sub> system, direct measurements extended from 77°K to 1124°K. Four different experimental techniques were employed; the Loschmidt, the Ney-Armistead, the point-source, and the chromatographic method. Molecular beam scattering data lead to diffusion coefficient values up to 10,000°K.

A graph of the  $\log D_{\text{He} - \text{N}_2}$  versus  $\log T$  ( $77^\circ\text{K} \leq T \leq 10,000^\circ\text{K}$ ) indicated that a correlation of the form  $D = D_0 T^S$  was applicable. Since measurements had been made at various composition, the experimental values of  $D_{\text{He} - \text{N}_2}$  were connected to a fixed composition by a semi-empirical correlation<sup>1</sup>. At a maximum the effect was taken to be 4% between helium trace in nitrogen and nitrogen trace in helium. Experimental data on composition dependence ranged from 6 to 2%. Data were reduced to nitrogen trace

1. Mason, E. A., S. Weissman and R. P. Wendt, Phys. Fluids 7, 174 (1964)

composition. The diffusion data were least squared with equal weights and in various temperature ranges. The correlated results for the He - N<sub>2</sub> system are presented in Table 1.

#### Ar - N<sub>2</sub>

The Ar - N<sub>2</sub> system was measured by only four independent scientist over a narrow temperature range,  $244 \leq T \leq 523^\circ\text{K}$ . Available scattering results were used to calculate diffusion coefficients at elevated temperatures. There exists an approximate 500°K gap between experimental data and the lower temperature limit for indirect determinations.

#### Ne - Ar

The Ne - Ar system received only one set of direct measurements in the temperature range of 273 to 318°K. Relative measurements are available down to 90°K. Diffusion coefficients calculated from viscosity data range from 72 to 523°K. It was found that analysis of thermal conductivity data yields only qualitatively correct diffusion coefficients, probably because the thermal conductivity measurements are not sufficiently accurate. Molecular beam results were used to calculate  $D_{\text{Ne - Ar}}$  at elevated temperatures. Agreement with the direct measurements is very good.

Recently, Leonas and coworkers at Moscow State University have carried out high-energy molecular beam scattering studies of all possible pairs of the noble gases (except radon). Agreement with the earlier MIT measurements is satisfactory for all systems measured in both laboratories (He - He, Ne - Ne, Ar - Ar, Kr - Kr, Xe - Xe, He - Ar, Ne - Ar). It is therefore now possible to obtain values of  $D_{12}$  for all possible noble gas pairs up to temperatures of the order of  $10^4$ °K. These calculations are in progress.

Future work will be the evaluation of all noble gas pair data.



Table 1: Correlation of Binary Dilute Gas Diffusion Coefficients at 1 atm. Pressure

System	Temperature Range, °K	Correlation Equations $D(x_1 = 0) = D_0 T^S$	Evaluation	
			av. als. %	RMS %
He - Ar	70-273	$4.65 \times 10^{-5} T^{1.694(a)}$	3.4	3.7
	273-1063	$3.53 \times 10^{-5} T^{1.743}$	1.7	2.2
	273-1063	$3.34 \times 10^{-5} T^{1.752(b)}$	1.8	2.3
He - N <sub>2</sub>	244-1124	$4.41 \times 10^{-5} T^{1.697(c)}$	1.7	2.2
Ar - N <sub>2</sub>	244-335	$4.85 \times 10^{-6} T^{1.862}$	0.9	1.2
Ne - Ar	193-523	$1.82 \times 10^{-5} T^{1.716}$	2.0	2.6

(a) results below 273°K were nomalized to a value of 0.623 cm<sup>2</sup>/sec at 273.15°K. This point was calculated from  $D = 3.53 \times 10^{-5} T^{1.743}$

(b) correlation was derived from least squares fit of the weighted data between 273 and 15000°K

(c) the one data point at 77°K lies 2.9% higher than the value calculated from this equation

Table II: Diffusion Coefficients Correlations at Elevated Temperatures

System	$D = D_0 T^S$	Temperature Range °K
He - Ar	$3.34 \times 10^{-5} T^{1.752}$	$1000 \leq T \leq 15000$
He - N <sub>2</sub>	$2.42 \times 10^{-5} T^{1.783}$	$1000 \leq T \leq 10000$
Ar - N <sub>2</sub>	$1.57 \times 10^{-5} T^{1.757}$	$1000 \leq T \leq 10000$
Ne - Ar	$1.85 \times 10^{-5} T^{1.718}$	$1000 \leq T \leq 15000$

BIBLIOGRAPHY OF DIFFUSION COEFFICIENTS  
FOR DILUTE BINARY NOBLE GAS MIXTURES

SYSTEM	AUTHOR(S)	REFERENCE
$\text{He}^3 - \text{He}^3$	Luszczynski, K., R. E. Norberg and J. E. Opfer	Phys. Rev. <u>128</u> , 186 (1962)
$\text{He}^3 - \text{He}^4$	Bendt, P. J.	Phys. Rev. <u>110</u> , 85 (1958)
*He - He	Amdur, I. and E. A. Mason	Phys. Fluids <u>1</u> , 370 (1958)
He - Ne	Srivastava, K. P. and A. K. Barua	Indian J. Phys. <u>33</u> , 229 (1959)
He - Ne	Wasik, S.	J. Chem. Phys. to be published
*He - Ne	Weissman, S. and E. A. Mason	J. Chem. Phys. <u>37</u> , 1289 (1962)
*He - Ne	Weissman, S.	<u>Adv. Thermophys.</u> <u>Prop. at Extreme</u> <u>Temp. and Pressure</u> , ASME, PP. 12-18 (1965)
He - Ar	Schmidt, R.	Ann. Physik. <u>14</u> , 801 (1904)
He - Ar	Lonius, A.	Ann. Physik. <u>29</u> , 664 (1909)
He - Ar	Strehlow, R. A.	J. Chem. Phys. <u>21</u> , 2101 (1953)
He - Ar	" Schafer, K. and H. Moesta	Z. Elektrochem. <u>58</u> , 743 (1954)
He - Ar	Saxena, S. C. and E. A. Mason	Mol. Phys. <u>2</u> , 379 (1959)
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He - Ar	Walker, R. E. and A. A. Westenberg	J. Chem. Phys. <u>31</u> , 519 (1959) and personal communication
He - Ar	Suetin, P. E. and B. A. Ivakin	Sov. Phys.-Tech. Phys. <u>6</u> , 359 (1961)
He - Ar	Giddings, J. C. and S. L. Seager	Ind. Eng. Chem. Fundamentals <u>1</u> , 277 (1962)
He - Ar	Suetin, P. E.	<u>Heat and Mass Transfer</u> , Vol. 1, <u>Thermophysical Characteristics of Materials and Methods for their Determination</u> , P. 188 (1962) TRANS. ORNL-TR-316 (1964)
He - Ar	Golubev, I. F. and A. G. Bondarenko	Gaz. Prom. <u>8</u> , 46 (1963)
He - Ar	Seager, S. L., L. R. Geerston and J. C. Giddings	J. Chem. and Eng. Data <u>8</u> , 168 (1963)
He - Ar	Holsen, J. N. and M. R. Strunk	Ind. Eng. Chem. Fundamentals <u>3</u> , 143 (1964)
He - Ar	Ivakin, B. A. and P. E. Suetin	Sov. Phys.-Tech. Phys. <u>9</u> , 866 (1964)
He - Ar	Ljunggren, S.	Arkiv. Kemi (Sweden) <u>24</u> , 1 (1965)

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He - Ar	Malinauskas, A. P.	J. Chem. Phys. <u>44</u> , (1966) Preprint
He - Ar	Wasik, S.	J. Chem. Phys., to be published
*He - Ar	Amdur, I. and E. A. Mason	Phys. Fluids <u>1</u> , 370 (1958)
*He - Ar	Weissman, S. and E. A. Mason	J. Chem. Phys. <u>37</u> , 1289 (1962)
*He - Ar	Weissman, S.	<u>Adv. in Thermophys. Prop. at Extreme Temp. and Pressure</u> , ASME, pp. 12-18 (1965)
*He - Ar	Mason, E. A. and F. J. Smith	J. Chem. Phys. <u>44</u> , 3100 (1966)
He - Kr	Srivastave, K. P. and A. K. Barua	Indian J. Phys. <u>33</u> , 229 (1959)
He - Kr	Durbin, L. and R. Kobayashi	J. Chem. Phys. <u>37</u> , 1643 (1962)
He - Kr	Srivastava, B. N. and R. Paul	Physica <u>28</u> , 646 (1962)
He - Kr	Watts, H.	Trans. Faraday Soc. <u>60</u> , 1745 (1964)
He - Kr	Malinauskas, A. P.	J. Chem. Phys. <u>44</u> , (1966) Preprint
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\*Calculation

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He - Xe	Srivastava, K. P.	Physica <u>25</u> , 571(1959)
He - Xe	Malinauskas, A. P.	J. Chem. Phys. <u>42</u> , 156 (1965)
He - Xe	Watts, H.	Can. J. Chem. <u>43</u> , 431 (1965)
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He - Rn	Hirst, W. and G. E. Harrison	Proc. Roy. Soc. (London) <u>A 169</u> , 573 (1939)
Ne - Ne	Groth, W. and E. Sussner	Z. Physik Chem. (Leipzig) <u>A 193</u> , 296 (1944)
Ne - Ne	Winn, E. B.	Phys. Rev. <u>80</u> , 1024 (1950)
*Ne - Ne	Amdur, I. and E. A. Mason	Phys. Fluids <u>1</u> , 370 (1958)
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Ne - Kr	Paul, R.	Indian J. Phys. <u>36</u> , 464 (1962)
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Ackermann, G.	Ing. Archiv. <u>5</u> , 124 (1934)	H <sub>2</sub> O - Air
Amdur, I., J. W. Irvine, Jr. E. A. Mason and J. Ross	J. Chem. Phys. <u>20</u> , 436 (1952)	CO <sub>2</sub> - CO <sub>2</sub> CO <sub>2</sub> - N <sub>2</sub> O
Amdur, I. and T. F. Schatzki	J. Chem. Phys. <u>27</u> , 1049 (1957)	Ar - Xe, Xe - Xe, Ar - Ar
Amdur, I. and E. A. Mason	Phys. Fluids <u>1</u> , 370 (1958)	He - He, He - Ar, Ne - Ne, Ar - Ar, Kr - Kr, Xe - Xe, N <sub>2</sub> - N <sub>2</sub>
Amdur, I. and T. F. Schatzki	J. Chem. Phys. <u>29</u> , 1425 (1958)	Ar - Xe
Amdur, I. and L. M. Shuler	J. Chem. Phys. <u>38</u> , 188 (1963)	CO - CO, CO - N <sub>2</sub>
Amdur, I. and J. W. Beatty, Jr.	J. Chem. Phys. <u>42</u> , 3361 (1965)	H <sub>2</sub> - TH, H <sub>2</sub> - T <sub>2</sub> , D <sub>2</sub> - TH, D <sub>2</sub> - T <sub>2</sub>
Amdur, I. and A. P. Malinauskas	J. Chem. Phys. <u>42</u> , 3355 (1965)	He - T <sub>2</sub> , He - TH
Anderson, L. W. and A. T. Ramsey	Phys. Rev. <u>132</u> , 712 (1963)	He - Na, Ne - Na
Andrew, S. P. S.	Chem. Eng. Sci. <u>4</u> , 269 (1955)	NH <sub>3</sub> - AIR, AIR - CO <sub>2</sub> , AIR - SO <sub>2</sub> , AIR - Cl <sub>2</sub> , AIR - Br <sub>2</sub>

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Arnold, J. H.	Trans. A.I.Ch.E. <u>40</u> , 361 (1944)	AIR - CS <sub>2</sub>
Barus, C.	Proc. Natl. Acad. Sci. U. S. <u>10</u> , 153 (1924)	COAL GAS - AIR
Barus, C.	Proc. Natl. Acad. Sci. U. S. <u>10</u> , 447 (1924)	H <sub>2</sub> - AIR
Baumgartner, G.	Sitzber., Akad. Wiss., Wien (II) <u>75</u> , 313 (1877)	H <sub>2</sub> - ALCOHOL, H <sub>2</sub> - CHCl <sub>3</sub> , H <sub>2</sub> - CS <sub>2</sub> , H <sub>2</sub> - SULPHURIC ACID, COAL GAS - ALCOHOL, COAL GAS - CHCl <sub>3</sub> , COAL GAS - CS <sub>2</sub> , COAL GAS - SULPHURIC ACID, AIR - ALCOHOL, AIR - CHCl <sub>3</sub> , AIR - CS <sub>2</sub> , AIR - SULPHURIC ACID, CO <sub>2</sub> - CHCl <sub>3</sub> , CO <sub>2</sub> - CS <sub>2</sub> , CO <sub>2</sub> - SULPHURIC ACID
Baumgartner, G.	Sitzber., Akad. Wiss., Wien (II) <u>75</u> , 679 (1877)	H <sub>2</sub> - CS <sub>2</sub> , COAL GAS - CS <sub>2</sub> , AIR - CS <sub>2</sub> , CO <sub>2</sub> - CS <sub>2</sub>
Bendt, P. J.	Phys. Rev. <u>110</u> , 85 (1958)	He <sup>3</sup> - He <sup>4</sup> , H <sub>2</sub> - D <sub>2</sub>
Bernheim, R. A.	J. Chem. Phys. <u>36</u> , 135 (1962)	He - Rb

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Bernheim, R. A. and M. W. Korte	J. Chem. Phys. <u>42</u> , 2721 (1965)	He - K
Birks, J. and R. S. Bradley	Proc. Roy. Soc. (London) <u>A162</u> , 511 (1937)	H <sub>2</sub> - Di - n - BUTYL PHTHALATE, AIR - Di - n - BUTYL PHTHALATE, FREON - 12 - (CCl <sub>2</sub> F <sub>2</sub> ) - Di - n - BUTYL PHTHALATE
Boardman, L. E. and N. E. Wild	Proc. Roy. Soc. (London) <u>A162</u> , 511 (1937)	H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub> , CO <sub>2</sub> - N <sub>2</sub> O
Bohemen, J. and J. H. Purnell	J. Chem. Soc., p. 360 (1961)	H <sub>2</sub> - N <sub>2</sub> , N <sub>2</sub> - O <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub>
Bondarenko, A. G. and I. F. Golubev	Gaz. Prom. <u>9</u> , 50 (1964) (In Russian), Trans. ORNL - Tr - 1074	H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub>
Bournia, A., J. Coull and G. Houghton	Proc. Roy. Soc. (London) <u>A261</u> , 227 (1961)	1,3 - BUTADIENE - 1 - BUTYNE
Boyd, C. A., N. Stein, V. Steingrimsson and W. F. Rumpel	J. Chem. Phys. <u>19</u> , 548 (1951)	H <sub>2</sub> - CH <sub>4</sub> , H <sub>2</sub> - C <sub>2</sub> H <sub>6</sub> , H <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - SF <sub>6</sub> , N <sub>2</sub> - C <sub>2</sub> H <sub>4</sub> , N <sub>2</sub> - C <sub>2</sub> H <sub>6</sub> , N <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - nC <sub>4</sub> H <sub>10</sub> , N <sub>2</sub> - iso C <sub>4</sub> H <sub>10</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System(s)
Bradley, R. S., M. G. Evans and R. W. Whytlaw-Gray	Proc. Roy. Soc. (London) <u>A186</u> , 368 (1946)	AIR - BUTYL STEARATE, AIR - DIBUTYL PHTHALATE
Bradley, R. S. and A. D. Shellard	Proc. Roy. Soc. (London) <u>A198</u> , 239 (1949)	AIR - n - C <sub>16</sub> H <sub>34</sub> , AIR - n - C <sub>17</sub> H <sub>36</sub> , AIR - n - C <sub>18</sub> H <sub>38</sub>
Braune, H. and F. Zehle	Z. Physik. Chem. <u>49B</u> , 247 (1941)	HCl - HCl, HBr - HBr
Breetveld, J. D., R. DiPippo and J. Kestin	J. Chem. Phys. <u>45</u> , 124 (1966)	Ne - CO <sub>2</sub>
Brockett, C. P.	J. Chem. Educ. <u>43</u> , 207 (1966)	AIR - Br <sub>2</sub>
Brookfield, K. J. et al.	Proc. Roy. Soc. (London) <u>A190</u> , 59 (1947)	H <sub>2</sub> O - AIR, AIR - FURFURAL, AIR - METHYL SALICYLATE
Browning, R. W. and J. W. Fox	Proc. Roy. Soc. (London) <u>A278</u> , 247 (1964)	H - H <sub>2</sub>
Bunde, R. E.	Univ. Wisconsin, Naval Research Laboratory Report CM - 850 (1955)	H <sub>2</sub> - He, H <sub>2</sub> - NH <sub>3</sub> , H <sub>2</sub> - N <sub>2</sub> , NH <sub>3</sub> - N <sub>2</sub>
Carmichael, L. T., H. H. Reamer, B. H. Sage and W. N. Lacey	Ind. Eng. Chem. <u>47</u> , 2205 (1955)	CH <sub>4</sub> - n - HEPTANE

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System(s)
Carmichael, L. T., B. H. Sage, and W. N. Lacey	Am. Doc. Inst., Doc. No. 4661 (1955)	CH <sub>4</sub> - n - HEXANE, C <sub>2</sub> H <sub>6</sub> - n - HEXANE, C <sub>3</sub> H <sub>8</sub> - n - HEXANE
Chang, G. T.	Thesis, Rice University (USA) 1966	He - N <sub>2</sub> , CH <sub>4</sub> - C <sub>2</sub> H <sub>6</sub> , CH <sub>4</sub> - C <sub>3</sub> H <sub>8</sub> , CH <sub>4</sub> - n - BUTANE
Clarke, J. K. and A. R. Ubbelohde	J. Chem. Soc., 2050 (1957)	n - HEPTANE - H <sub>2</sub> , - D <sub>2</sub> , - He, - N <sub>2</sub> , - Ar, n - OCTANE - H <sub>2</sub> , - D <sub>2</sub> , - He, - N <sub>2</sub> , - Ar, 2:2:4 - TRIMETHYLPENTANE - H <sub>2</sub> , - D <sub>2</sub> , - He, - N <sub>2</sub> , - Ar, 2:4 - DIMETHYLPENTANE - H <sub>2</sub> , - D <sub>2</sub> , - He, - N <sub>2</sub> , - Ar
Cordes, H. and K. Kerl	Z. Physik. Chem. (Frankfurt) <u>45</u> , 369 (1965)	H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - Ar
Coulliette, J. H.	Phys. Rev. <u>32</u> , 636 (1928)	Hg - Hg
Crider, W. S.	J. Am. Chem. Soc. <u>78</u> , 924 (1956)	H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O - N <sub>2</sub> , H <sub>2</sub> O - CO <sub>2</sub>
Cummings, G. A. McD. and A. R. Ubbelohde	J. Chem. Soc., 3751 (1953)	n - HEXANE - H <sub>2</sub> , - N <sub>2</sub> , - O <sub>2</sub> , - Ar, 2:3 - DIMETHYLBUTANE - H <sub>2</sub> , - N <sub>2</sub> , - O <sub>2</sub> , - Ar, CYCLOHEXANE - H <sub>2</sub> , - N <sub>2</sub> , - O <sub>2</sub> , - Ar



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Author(s)	Reference	System(s)
Cummings, G. A. McD. and A. R. Ubbelohde - Cont.		METHYLCYCLOPENTANE - H <sub>2</sub> , - N <sub>2</sub> , - O <sub>2</sub> , - Ar, n - OCTANE - H <sub>2</sub> , - N <sub>2</sub> , - O <sub>2</sub> , - Ar, 2:2:4 - TRIMETHYLPEN- TANE - H <sub>2</sub> , - N <sub>2</sub> , - O <sub>2</sub> , - Ar, n - DECANE - H <sub>2</sub> , - N <sub>2</sub> 2:3:3 - TRIMETHYLHEPTANE - H <sub>2</sub> , - N <sub>2</sub> , n - DODECANE - H <sub>2</sub> , - N <sub>2</sub>
Cummings, G. A. McD., E. McLaughlin and A. R. Ubbelohde	J. Chem. Soc., 1141 (1955)	HEXA - 1:5 - DIENE - H <sub>2</sub> , - N <sub>2</sub> , 2:3 DIMETHYLBUTA - 1:3 - diene - H <sub>2</sub> , - N <sub>2</sub> , 2:3 - DIMETHYLBUT - 2 - ene - H <sub>2</sub> , - N <sub>2</sub> , n - HEPTANE - H <sub>2</sub> , - N <sub>2</sub> , n - NONANE - H <sub>2</sub> , - N <sub>2</sub>
Cvetanovic, R. J. and D. J. LeRoy	J. Chem. Phys. 20, 343 (1952)	Na - N <sub>2</sub>
DeLucca, L. B.	Phys. Rev. 95, 306A (1954)	N <sub>2</sub> - N <sub>2</sub>
Durbin, L. and R. Kobayashi	J. Chem. Phys. 37, 1643 (1962)	He - Kr, C <sub>2</sub> H <sub>4</sub> - Kr, N <sub>2</sub> - Kr, Ar - Kr, CO <sub>2</sub> - Kr - Kr
Ember, G., J. R. Ferron and K. Wohl	J. Chem. Phys. 37, 891 (1962)	CO <sub>2</sub> - CO <sub>2</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author(s)	Reference	System(s)
Ember, G., J. R. Ferron and K. Wohl	A.I.Ch.E. J. <u>10</u> , 68 (1964)	CH <sub>4</sub> - CH <sub>4</sub> , CH <sub>4</sub> - CO <sub>2</sub> , CO <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> O - CO <sub>2</sub>
Evans, E. V. and C. N. Kenney	Proc. Roy. Soc. (London) <u>A284</u> , 540 (1965)	H <sub>2</sub> - Ar, H <sub>2</sub> - SF <sub>6</sub> , He - SF <sub>6</sub> , C <sub>2</sub> H <sub>4</sub> - N <sub>2</sub> , N <sub>2</sub> - SF <sub>6</sub> , Ar - SF <sub>6</sub>
Fairbanks, D. F. and C. R. Wilke	Ind. Eng. Chem. <u>42</u> , 471 (1950)	H <sub>2</sub> - TOLUENE, H <sub>2</sub> - ETHYL PROPIONATE, AIR - TOLUENE, AIR - ETHYL PROPIONATE
Franzen, W.	Phys. Rev. <u>115</u> , 850 (1959)	Ne - Rb, Ar - Rb
Gaede, W.	Ann. Physik <u>46</u> , 357 (1915)	H <sub>2</sub> - Hg, AIR - Hg
Giddings, J. C. and S. L. Seager	J. Chem. Phys. <u>33</u> , 1579 (1960)	H <sub>2</sub> - N <sub>2</sub>
Giddings, J. C. and S. L. Seager	Ind. Eng. Chem. Fundamentals <u>1</u> , 277 (1962)	H <sub>2</sub> - He, H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , He - NH <sub>3</sub> , He - N <sub>2</sub> , He - O <sub>2</sub> , He - Ar, N <sub>2</sub> - CO <sub>2</sub>
Gilliland, E. R.	Ind. Eng. Chem. <u>26</u> , 681 (1934)	AIR - H <sub>2</sub> O, n - BUTYL ALCOHOL, TOLUENE, sec BUTYL ALCOHOL, ETHYL ACETATE, CHLOROBENZENE, ANILINE, sec AMYL ALCOHOL, DIPHENYL, ISOPROPYL ALCOHOL

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System(s)
Golubev, I. F. and A. G. Bondarenko	Gaz. Prom. <u>9</u> , 46 (1963) (In Russian)	H <sub>2</sub> - Ar, He - Ar
Goryunova, N. A. and E. V. Kuvshinski	Zh. Techn. Fiz. (USSR) <u>18</u> , 1421 (1948)	AIR - ACETONE, CHLOROFORM, CYCLOHEXANE
Griboiedov, S.	J. Russ. Phys-Chem Soc. <u>25</u> , 36 (1893) (In Russian)	AIR - BENZENE, ETHYL ETHER, methyl formate, methyl acetate, methyl propionate
Grieverson, P. and E. T. Turkdogan	J. Phys. Chem. <u>68</u> , 1547 (1964)	Ar - Cr, Ar - Fe, Ar - Co, Ar - Ni
Groth, W. and P. Harteck	Z. Elektrochem. <u>47</u> , 167 (1941)	Xe - Xe, Kr - Kr
Groth, W. and E. Sussner	Z. Physik. Chem. (Leipzig) <u>A193</u> , 296 (1944)	Ne - Ne
Groth, W. and P. Harteck	Z. Physik. Chem. (Leipzig) <u>199</u> , 114 (1952)	H <sub>2</sub> - D <sub>2</sub> , D <sub>2</sub> - He, D <sub>2</sub> - Ne, D <sub>2</sub> - Ar, D <sub>2</sub> - Kr, D <sub>2</sub> - Xe
Guglielmo, G.	Atti Accad. Torino <u>17</u> , 54 (1881)	H <sub>2</sub> O - AIR
Guglielmo, G.	Atti Accad. Torino <u>18</u> , 93 (1882) (In Italian)	H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O - AIR, H <sub>2</sub> O - CO <sub>2</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author(s)	Reference	System(s)
Gush, L. L.	Trans. Inst. Chem. Engrs. <u>26</u> , 142, (1948)	AIR - ACETONE
Harteck, P., and H. W. Schmidt	Z. Physik Chem. (Leipzig) <u>B21</u> , 447 (1933)	H <sub>2</sub> - H <sub>2</sub>
Hartland, A. and M. Lipsicas	Phys. Letters <u>3</u> , 212 (1963)	H <sub>2</sub> - H <sub>2</sub>
Heath, H. R., T. L. Ibbs and N. E. Wild	Proc. Roy. Soc. (London) <u>A178</u> , 380 (1941)	H <sub>2</sub> - D <sub>2</sub>
Heinzelmann, F. J., D. T. Wasan and C. R. Wilke	Ind. Eng. Chem. Fundamentals <u>4</u> , 55 (1965)	AIR - BENZENE
van der Held, E. F. M. and M. Miesowicz	Physica <u>4</u> , 559 (1937)	N <sub>2</sub> - Na
Hippenmeyer, B.	Z. Angew. Phys. <u>1</u> , 549 (1949)	H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O - N <sub>2</sub>
Hirst, W. and G. E. Harrison	Proc. Roy. Soc. (London) <u>A169</u> , 573 (1939)	H <sub>2</sub> - Rn, He - Rn, Ne - Rn, AIR - Rn, Ar - Rn
Holsen, J. N. and M. R. Strunk	Ind. Eng. Chem. Fundamentals <u>3</u> , 143 (1964)	He - AIR, He - Ar, He - CO <sub>2</sub> , AIR - CO <sub>2</sub> , Ar - CO <sub>2</sub>
Houghton, H. G.	J. Appl. Phys. <u>4</u> , 419 (1933)	H <sub>2</sub> O - AIR (At various humidities)

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Author (s)	Reference	System (s)
Huber, J. F. K., and G. van Vught	Ber. Bunsenges Physik. Chem. <u>69</u> , 821 (1965)	N <sub>2</sub> - Hexane
Hudson, G. H., J. C. McCoubrey and A. R. Ubbelohde	Trans. Faraday Soc. <u>56</u> , 1144 (1960)	H <sub>2</sub> - BENZENE, CYCLOHEXANE, PYRIDINE, PIPERIDINE, THIOPHEN, TETRAHYDROTHIOPHEN N <sub>2</sub> - BENZENE, CYCLOHEXANE, PYRIDINE, PIPERIDINE, THIOPHEN, TETRAHYDROTHIOPHEN, O <sub>2</sub> - BENZENE, CYCLOHEXANE, PYRIDINE, PIPERIDINE, THIOPHEN, TETRAHYDROTHIOPHEN
Hutchinson, F.	Phys. Rev. <u>72</u> , 1256 (1947)	Ar - Ar
Hutchinson, F.	J. Chem. Phys. <u>17</u> , 1081 (1949)	Ar - Ar
van Itterbeek, A. and J. Nihoul	Acustica <u>7</u> , 180 (1957)	H <sub>2</sub> - He, H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - O <sub>2</sub>
Ivakin, B. A. and P. E. Suetin	Zh. Techn. Fiz. <u>33</u> , 1007 (1963) Trans. Sov. Phys.- <u>8</u> , 748 (1964)	H <sub>2</sub> - D <sub>2</sub> , H <sub>2</sub> - CO, H <sub>2</sub> - NH <sub>3</sub> , D <sub>2</sub> - He, D <sub>2</sub> - NH <sub>3</sub> , D <sub>2</sub> - CO, D <sub>2</sub> - N <sub>2</sub> , D <sub>2</sub> - AIR, D <sub>2</sub> - Ar, D <sub>2</sub> - CO <sub>2</sub> , D <sub>2</sub> - SF <sub>6</sub> , He - NH <sub>3</sub> , He - CO, NH <sub>3</sub> - CO, NH <sub>3</sub> - SF <sub>6</sub> , CO - N <sub>2</sub> , CO - Ar, CO - CO <sub>2</sub> , CO - SF <sub>6</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author(s)	Reference	System(s)
Ivakin, B. A. and P. E. Suetin	Zh. Techn. Fiz. <u>34</u> , 1115 (1964) (In Russian) Trans. Sov. Phys.-Tech. Phys. <u>9</u> , 866 (1964)	H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - CO, H <sub>2</sub> - Ar, H <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - SF <sub>6</sub> , He - N <sub>2</sub> , He - CO, He - AIR, He - AIR, He - Ar, He - CO <sub>2</sub> , He - SF <sub>6</sub> , N <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - SF <sub>6</sub> , Ar - CO <sub>2</sub> , Ar - SF <sub>6</sub> , CO <sub>2</sub> - SF <sub>6</sub>
Jona, F.	J. Chem. Phys. <u>42</u> , 1025 (1965)	GeBr <sub>2</sub> - GeBr <sub>4</sub> , GeI <sub>2</sub> - GeI <sub>4</sub>
Jorgensen, F. and H. Watts	Chem. and Ind. (London) p. 1440 (1961)	AIR - BENZENE
Kimpton, D. D. and F. T. Wall	J. Phys. Chem. <u>56</u> , 715 (1952)	CH <sub>4</sub> - H <sub>2</sub> O, H <sub>2</sub> O - C <sub>2</sub> H <sub>4</sub> , H <sub>2</sub> O - AIR, H <sub>2</sub> O - C <sub>2</sub> H <sub>6</sub> , H <sub>2</sub> O - C <sub>3</sub> H <sub>8</sub> , H <sub>2</sub> O - SO <sub>2</sub> , D <sub>2</sub> O - AIR
Klibanov, Ts. M., and V. V. Pomerantsev, and D. A. Frank- Kamenetsky	Zh. Techn. Fiz. (USSR) 12, 14 (1942) (In Russian)	H <sub>2</sub> O - AIR, AIR - CO <sub>2</sub>
Klotz, I. M. and D. K. Miller	J. Am. Chem. Soc. <u>69</u> , 2557 (1947)	HCN - AIR, AIR - CNCl, AIR - COCl <sub>2</sub> , AIR - CCl <sub>3</sub> NO <sub>2</sub>
Knox, J. H. and L. McLaren	Anal. Chem. <u>36</u> , 1477 (1964)	ETHYLENE - N <sub>2</sub>
Knox, J. H. and L. McLaren	Anal. Chem. <u>35</u> , 449 (1963)	N <sub>2</sub> - C <sub>2</sub> H <sub>4</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Kohn, J. P. and N. Romero	J. Chem. Eng. Data <u>10</u> , 125 (1965)	CH <sub>4</sub> - n - C <sub>6</sub> H <sub>14</sub> , CH <sub>4</sub> - 3 - METHYLPENTANE
Kosov, N. D.	Issledovanie Fiz. Osnov. Rabochego Protsessa Topoki Pechei (Alma-Atai Akad. Nauk Kazakh. S. S. R.) Sbornik, 285 (1957) ORNL- tr-1073	AIR - H <sub>2</sub> , AIR - CO <sub>2</sub> , AIR - Cl <sub>2</sub> , AIR - CH <sub>2</sub> CH <sub>2</sub> ,
Krongelb, S. and M. W. P. Strandberg	J. Chem. Phys. <u>31</u> , 1196 (1959)	O - O <sub>2</sub>
Langmuir, I.	Phys. Rev. <u>12</u> , 368 (1918)	AIR - I <sub>2</sub>
LeBlanc, M. and G. Wupperman	Z. Physik. Chem. (Leipzig) <u>91</u> , 143 (1916) (In German)	H <sub>2</sub> O - AIR, AIR - BENZENE, AIR - PROPYLACETATE, AIR - CHLORBENZENE
Lee, C. Y. and C. R. Wilke	Ind. Eng. Chem. <u>46</u> , 2381 (1954)	He - H <sub>2</sub> O, He - ETHYL ALCOHOL He - BENZENE, He - NITROBENZENE, H <sub>2</sub> O - AIR, AIR - ETHYL ALCOHOL, AIR - BENZENE, AIR - NITROBENZENE, H <sub>2</sub> O - FREON - 12, ETHYL ALCOHOL - FREON - 12, FREON - 12 - BENZENE

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Author (s)	Reference	System(s)
Legowski, S.	J. Chem. Phys. <u>41</u> , 1313 (1964)	He - Cs, Ne - Cs, Ar - Cs
Lipsicas, M.	J. Chem. Phys. <u>36</u> , 1235 (1962)	H <sub>2</sub> - H <sub>2</sub>
Ljunggren, S.	Arkiv. Kemi (Sweden) <u>24</u> , 1 (1965)	H <sub>2</sub> - UF <sub>6</sub> , He - Ar, He - UF <sub>6</sub> Ne - UF <sub>6</sub> , N <sub>2</sub> - UF <sub>6</sub> , O <sub>2</sub> - UF <sub>6</sub> , Ar - UF <sub>6</sub> , Kr - UF <sub>6</sub> , Xe - UF <sub>6</sub>
Lonius, A.	Ann. Physik <u>29</u> , 664 (1909) (In German)	He - Ar, H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - O <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - O <sub>2</sub>
Lonsdale, H. K. and E. A. Mason	J. Phys. Chem. <u>61</u> , 1544 (1957)	H <sub>2</sub> - CO <sub>2</sub> , He - CO <sub>2</sub>
Loschmidt, J.	Sitzber., Akad. Wiss., Wien <u>61</u> , 367 (1870) (In German)	H <sub>2</sub> - O <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , AIR - CO <sub>2</sub>
Loschmidt, J.	Sitzber., Akad. Wiss., Wien <u>62</u> , 468 (1870) (In German)	H <sub>2</sub> - O <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - SO <sub>2</sub> , CH <sub>4</sub> - CO <sub>2</sub> , CO - O <sub>2</sub> , CO - CO <sub>2</sub> , AIR - CO <sub>2</sub> , O <sub>2</sub> - CO <sub>2</sub> , CO <sub>2</sub> - N <sub>2</sub> O
Luszczynski, K., R. E. Norberg and J. E. Opfer	Phys. Rev. <u>128</u> 186 (1962)	He <sup>3</sup> - He <sup>3</sup>



BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author(s)	Reference	System(s)
McCoubrey, A. O.	Phys. Rev. <u>93</u> 1249 (1954)	Hg <sub>2</sub> - Hg <sub>2</sub>
McCoubrey, A. O. and C. G. Matland	Phys. Rev. <u>101</u> 603 (1956)	Hg <sub>2</sub> - Hg <sub>2</sub>
McMurtie, R. L. and F. G. Keyes	J. Am. Chem. Soc. <u>70</u> , 3755 (1948)	H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O <sub>2</sub> - AIR
Mache, H.	Sitzber, Akad. Wiss., Wien <u>119</u> , 1399 (1910) (In German)	H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O - AIR
Mack, E., Jr.	J. Am. Chem. Soc. <u>47</u> , 2468 (1925)	AIR - I <sub>2</sub> , TOLUENE, NAPHTHALENE, DIPHENYL, BENZIDINE, n - OCTANE, ANILINE
Mackenzie, J. E. and H. W. Melville	Proc. Roy. Soc. (Edinburgh) <u>52</u> , 337 (1932)	H <sub>2</sub> - Br <sub>2</sub> , N <sub>2</sub> - Br <sub>2</sub> , O <sub>2</sub> - Br <sub>2</sub> , CO <sub>2</sub> - Br <sub>2</sub>
Mackenzie, J. E. and H. W. Melville	Proc. Roy. Soc. (Edinburgh) <u>53</u> , 255 (1933)	H <sub>2</sub> - Br <sub>2</sub> , CH <sub>4</sub> - Br <sub>2</sub> , HCl - Br <sub>2</sub> , Ar - Br <sub>2</sub> , CO <sub>2</sub> - Br <sub>2</sub> , N <sub>2</sub> O - Br <sub>2</sub>
McNeal, J.	J. Chem. Phys. <u>37</u> , 2726 (1962)	Rb - H <sub>2</sub> , - N <sub>2</sub> , - CH <sub>4</sub> , - C <sub>2</sub> H <sub>4</sub> , - C <sub>2</sub> H <sub>6</sub> , - C <sub>6</sub> H <sub>12</sub>
Maitland, C. G. and A. O. McCoubrey	Phys. Rev. <u>98</u> , 558 (1955)	Hg <sub>2</sub> - Hg <sub>2</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Malinauskas, A. P.	J. Chem. Phys. <u>42</u> , 156 (1965)	He - Ar, He - Xe, Ar - Xe
Malinauskas, A. P.	J. Chem. Phys. <u>44</u> , (1966) preprint	He - Ar, He - Kr, Ar - Kr, Kr - Xe
Mason, E. A., S. Weissman and R. P. Wendt	Phys. Fluids <u>7</u> , 174 (1964)	H <sub>2</sub> - Ar, T <sub>2</sub> - N <sub>2</sub> , T <sub>2</sub> - Ar, T <sub>2</sub> - CO <sub>2</sub>
Mason, E. A., B. K. Annis and M. Islam	J. Chem. Phys. <u>42</u> 3364 (1965)	H <sub>2</sub> - T <sub>2</sub> , D <sub>2</sub> - T <sub>2</sub>
Mason, E. A. and F. J. Smith	J. Chem. Phys. <u>44</u> 3100 (1966)	He - Ar, He - Kr
Mehta, V. D.	M. Sc. (TECH) Thesis, Bombay Univ. (1966)	H <sub>2</sub> - TRIETHYLAMINE, N <sub>2</sub> - ACETONE, N <sub>2</sub> - n - BUTYLAMINE N <sub>2</sub> - TRIETHYLAMINE, FREON - 12 - TRIETHYLAMINE
Miller, L. and P. C. Carman	Trans. Faraday Soc. <u>57</u> , 2143 (1961)	H <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - Kr, H <sub>2</sub> - CF <sub>2</sub> Cl <sub>2</sub> , CF <sub>2</sub> Cl <sub>2</sub> - CF <sub>2</sub> Cl <sub>2</sub> , CO <sub>2</sub> - CO <sub>2</sub> , Kr - Kr

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author(s)	Reference	System(s)
Miller, L. and P. C. Carman	Trans. Faraday Soc. <u>60</u> , 33 (1964)	H <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - Kr, H <sub>2</sub> - CF <sub>2</sub> Cl <sub>2</sub> , H <sub>2</sub> - Xe
Morgan, J. E. and H. I. Schiff	Can. J. Chem. <u>42</u> , 2300 (1964)	N - N <sub>2</sub> , O - N <sub>2</sub> , O - O <sub>2</sub> , O - Ar
Mueller, C. R. and R. W. Cahill	J. Chem. Phys. <u>40</u> , 651 (1964)	CH <sub>4</sub> - CH <sub>4</sub> , CH <sub>4</sub> - CO, CH <sub>4</sub> - N <sub>2</sub> , CH <sub>4</sub> - CF <sub>4</sub> , CHCH - CHCH, CHCH - CH <sub>3</sub> CH <sub>3</sub> , CH <sub>2</sub> CH - CH <sub>2</sub> CH <sub>2</sub> , CH <sub>2</sub> CH <sub>2</sub> - N <sub>2</sub> , CH <sub>2</sub> CH <sub>2</sub> - CH <sub>3</sub> CH <sub>3</sub> , CH <sub>3</sub> CH <sub>3</sub> - CH <sub>3</sub> CH <sub>3</sub>
Mullaly, J. M. and H. Jacques	Phil. Mag. <u>48</u> , 1105 (1924)	N <sub>2</sub> - Hg, Hg - I <sub>2</sub>
Naccari, A.	Nuovo Cimento <u>19</u> , 52 (1910) (In Italian)	ETHER - AIR
Nelson, E. T.	J. Appl. Chem. <u>6</u> , 286 (1956)	H <sub>2</sub> O - AIR, H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O - N <sub>2</sub> , H <sub>2</sub> O - COAL GAS, BENZENE - COAL GAS
Nettley, P. T.	Proc. Phys. Soc. (London) <u>B67</u> , 753 (1954)	H <sub>2</sub> - N <sub>2</sub>
Ney, E. P. and F. C. Armistead	Phys. Rev. <u>71</u> , 14 (1947)	UF <sub>6</sub> - UF <sub>6</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System(s)
Nikolaev, G. I. and V. B. Aleksovskii	Zh. Techn. Fiz. <u>34</u> , 753 (1964) Trans. Sov. Phys.- Tech. Phys. <u>9</u> , 575 (1964)	Ar - Zn
Obermayer, A.	Sitzber., Akad. Wiss., Wien <u>81</u> , 1102 (1880) (In German)	H <sub>2</sub> - O <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , AIR - CO <sub>2</sub> , N <sub>2</sub> - O <sub>2</sub> , CO <sub>2</sub> - N <sub>2</sub> O
Obermayer, A.	Sitzber., Akad. Wiss., Wien <u>87</u> , 188 (1883) (In German)	H <sub>2</sub> - CH <sub>4</sub> , H <sub>2</sub> - C <sub>2</sub> H <sub>4</sub> , H <sub>2</sub> - AIR, H <sub>2</sub> - C <sub>2</sub> H <sub>6</sub> , H <sub>2</sub> - CO, H <sub>2</sub> - O <sub>2</sub> , H <sub>2</sub> - N <sub>2</sub> O, H <sub>2</sub> - CO <sub>2</sub> , CO - O <sub>2</sub> , CO - C <sub>2</sub> H <sub>4</sub> , AIR - CO <sub>2</sub>
Obermayer, A.	Sitzber., Akad. Wiss., Wien <u>85</u> , 147 (1882) (In German)	H <sub>2</sub> - CO <sub>2</sub> , AIR - CO <sub>2</sub> , O <sub>2</sub> - CO <sub>2</sub>
Obermayer, A.	Sitzber., Akad. Wiss., Wien <u>85</u> , 738 (1882) (In German)	AIR - O <sub>2</sub> , N <sub>2</sub> - O <sub>2</sub> , AIR - CO <sub>2</sub> , N <sub>2</sub> O - CO <sub>2</sub>
Obermayer, A.	Sitzber., Akad. Wiss., Wien <u>96</u> , 546 (1887) (In German)	CH <sub>4</sub> - CO <sub>2</sub> , N <sub>2</sub> - O <sub>2</sub> , CO - CO <sub>2</sub> , C <sub>2</sub> H <sub>4</sub> - CO <sub>2</sub> , AIR - O <sub>2</sub> , AIR - CO <sub>2</sub>
Pakurar, T. A. and J. R. Ferron	preprint	N <sub>2</sub> - CO <sub>2</sub> , Ar - CO <sub>2</sub> , CO <sub>2</sub> - CO <sub>2</sub>
Pakurar, T. A. and J. R. Ferron	J. Chem. Phys. to be published (Preprint)	N <sub>2</sub> - CO <sub>2</sub> , Ar - CO <sub>2</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System(s)
Pakurar, T. I. and J. R. Ferron	J. Chem. Phys. <u>43</u> , 2917 (1965)	CO <sub>2</sub> - CO <sub>2</sub>
Parker, A. S., and H. C. Hottel	Ind. Eng. Chem. <u>28</u> , 1334 (1936)	N <sub>2</sub> - O <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub>
Paul, R. and I. B. Srivastava	Indian J. Phys. <u>35</u> , 465 (1961)	He - O <sub>2</sub> , O <sub>2</sub> - Ar, O <sub>2</sub> - Xe
Paul, R. and I. B. Srivastava	Indian J. Phys. <u>35</u> , 523 (1961)	He - N <sub>2</sub> , N <sub>2</sub> - Ar, N <sub>2</sub> - Xe
Paul, R. and I. B. Srivastava	J. Chem. Phys. <u>35</u> , 1621 (1961)	H <sub>2</sub> - Ne, H <sub>2</sub> - Ar, H <sub>2</sub> - Xe
Paul, R.	Indian J. Phys. <u>36</u> , 464 (1962)	Ne - Ke, Ar - Kr, Kr - Kr
Petit, M.-C.	Compt. Rend. <u>260</u> , 1368 (1965) (In French)	H <sub>2</sub> O - AIR
Pochettino, A.	Nuovo Cimento <u>8</u> , 5 (1914)	AIR - acetic acid, - methyl, formate, - propionic acid, methyl acetate, - ethyl acetate, - butyric acid, - i - Butyric acid, - methyl propionate, - ethyl acetate, - propyl formate, - Valeric acid, - i - valeric acid,

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Pochettino, A.	Cont.	<p>methyl butyrate, methyl -  i - Butyrate, - ethyl  propionate, propyl acetate,  - i - Butyl formate, -  caprylic acid, i - caprylic  acid, methyl valerate,  ethyl butyrate, ethyl - i -  butyrate, - propyl  propionate, butyl acetate,  - i - butyl acetate, - amyl  formate, i - amyl formate,  ethyl valerate, - propyl  valerate, propyl i -  butyrate, i - propyl i -  butyrate, i-butyl propionate,  propyl valerate, i - butyl  butyrate, i - butyl i-  butyrate, amyl propionate  i - butyl valerate, amyl  butyrate, amyl i - butyrate  propyl alcohol, i - propyl  alcohol, n - propyl bromide,  i - propyl bromide, n -  propyl iodide, i - propyl  iodide, ethyl ether, butyl  alcohol, i - butyl alcohol  safrole, i - safrole,  eugenol, i - eugenol,</p>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Pochettino, A. Cont.		butylamine, i - butylamine, diethylamine, propylbenzene, , i - propylbenzene, mesitylene, ethyl benzene, o - xylene, p - xylene benzyl chloride, o - chlorotoulene, m - chlorotoulene, p - chlorotoulene
Ramsey, A. T. and L. E. Anderson	Nuovo Cimento <u>32</u> , 1151 (1964)	H <sub>2</sub> - Na, He - Na, Na - N <sub>2</sub>
Raw, C. J. G.	J. Chem. Phys. <u>23</u> , 973 (1955)	BF <sub>3</sub> - CCl <sub>4</sub> , BCl <sub>3</sub> - CCl <sub>4</sub>
Raw, C. J. G. and H. Tang	J. Chem. Phys. <u>39</u> , 2616 (1963)	CF <sub>4</sub> - SF <sub>6</sub>
Reichenbacher, W. P. Muller, and A. Klemm	Z. Naturforsch. <u>20a</u> , 1529 (1965) (In German)	H <sub>2</sub> - HT, H <sub>2</sub> - DT, H <sub>2</sub> - T <sub>2</sub> , D <sub>2</sub> - HT, D <sub>2</sub> - DT, D <sub>2</sub> - T <sub>2</sub>
Richardson, J. F.	Chem. Eng. Sci. <u>10</u> , 234 (1959)	H <sub>2</sub> O - AIR, AIR - ACETONE, AIR - CCl <sub>4</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Rossie, K.	Forsch. Gebiete Ingenieurw <u>19B</u> , 49 (1953) (In German)	H <sub>2</sub> O - AIR
Rumpel, W. F.	Univ. of Wisconsin, Naval Research Laboratory Report, CM-851 (1955)	H <sub>2</sub> - He, He - N <sub>2</sub>
Rutherford, E. and H. T. Brooks	Trans. Roy. Soc. Canada <u>7</u> , 21 (1901)	Ra - AIR
Saxena, S. C. and E. A. Mason	Mol. Phys. <u>2</u> , 379 (1959)	H <sub>2</sub> - CO <sub>2</sub> , He - Ar, He - CO <sub>2</sub> , D <sub>2</sub> - CO <sub>2</sub>
<sup>m</sup> Schafer, K., H. Corte and H. Moesta	Z. Elektrochem. <u>55</u> , 662 (1951) (In German)	H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub>
<sup>v</sup> Schafer, K. and H. Moesta	Z. Elektrochem. <u>58</u> , 743 (1954)	H <sub>2</sub> - CO <sub>2</sub> , He - Ar, H <sub>2</sub> - N <sub>2</sub> , N <sub>2</sub> - Ar
<sup>m</sup> Schafer, K. and K. Schuhmann	Z. Elektrochem. <u>61</u> , 246 (1957) (In German)	Ne - Ar, Ar - Kr, Kr - Kr
<sup>n</sup> Schafer, K.	Z. Electrochem. <u>63</u> , 111 (1959) (In German)	H <sub>2</sub> - NH <sub>3</sub> , H <sub>2</sub> - SO <sub>2</sub> , N <sub>2</sub> - SO <sub>2</sub> Ar - SO <sub>2</sub> , CO <sub>2</sub> - SO <sub>2</sub>
<sup>n</sup> Schafer, K. and P. Reinhard	Z. Naturforsch. <u>18a</u> , 187 (1963) (In German)	CO <sub>2</sub> - CO <sub>2</sub>



BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System(s)
Schirmer, R.	Z. Ver. Deut Ing. Beihft Folge, 170 (1938)	WATER - AIR
Schmidt, R.	Ann. Physik <u>14</u> , 801 (1904) (In German)	H <sub>2</sub> - CO <sub>2</sub> , He - Ar
Schwartz, F. A. and J. E. Brow	J. Chem. Phys. <u>19</u> , 640 (1951)	H <sub>2</sub> - H <sub>2</sub> O, He - H <sub>2</sub> O, CH <sub>4</sub> - H <sub>2</sub> O, H <sub>2</sub> O - C <sub>2</sub> H <sub>4</sub> , H <sub>2</sub> O - N <sub>2</sub> , H <sub>2</sub> O - O <sub>2</sub> , H <sub>2</sub> O - CO <sub>2</sub>
Scott, D. S. and K. E. Cox	Can. J. Chem. Eng. <u>38</u> , 201 (1960)	H <sub>2</sub> - NH <sub>3</sub> , H <sub>2</sub> - N <sub>2</sub>
Seager, S. L., L. R. Geerston and J. C. Giddings	J. Chem. and Eng. Data <u>8</u> , 168 (1963)	He - N <sub>2</sub> , O <sub>2</sub> , Ar, CO <sub>2</sub> , METHANOL, ETHANOL, 1 - PROPANOL, 1 - BUTANOL, BENZENE, 1 - PENTANOL, 1 - HEXANOL
Spier, J. L.	Physica <u>6</u> , 453 (1939)	H <sub>2</sub> - Hg
Spier, J. L.	Physica <u>7</u> , 381 (1940)	N <sub>2</sub> - Cd, N <sub>2</sub> - Hg
Srivastava, B. N. and K. P. Srivastava	J. Chem. Phys. <u>30</u> , 984 (1959)	Ne - Ar, Ne - Kr, Ar - Kr
Srivastava, B. N. and R. Paul	Physica <u>28</u> , 646 (1962)	He - Kr, Kr - Kr

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Srivastava, B. N. and I. B. Srivastava	J. Chem. Phys. <u>36</u> , 2616 (1962)	NH <sub>3</sub> - Ar, NH <sub>3</sub> - Kr
Srivastava, B. N. and I. B. Srivastava	J. Chem. Phys. <u>38</u> , 1183 (1963)	NH <sub>3</sub> - (C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub> O
Srivastava, B. N. and A. Saran	Physica <u>32</u> , 110 (1966)	ACETONE - Kr, Kr - CHLOROFORM
Srivastava, I. B.	Indian J. Phys. <u>36</u> , 193 (1962)	He - NH <sub>3</sub> , NH <sub>3</sub> - Ne, NH <sub>3</sub> - Xe
Srivastava, K. P.	Physica <u>25</u> , 571 (1959)	He - Xe, He - Ar, Xe - Ar
Srivastava, K. P. and A. K. Barua	Indian J. Phys. <u>33</u> , 229 (1959)	He - Ne, He - Kr, Ne - Xe
Stefan, J.	Sitzber., Akad. Wiss., Wien <u>63</u> , 63 (1871) (In German)	H <sub>2</sub> O - AIR
Stefan, J.	Sitzber., Akad. Wiss., Wien <u>68</u> , 385 (1873) (In German)	AIR - ETHER, AIR - CS <sub>2</sub>
Strehlow, R. A.	J. Chem. Phys., <u>21</u> , 2101 (1953)	H <sub>2</sub> - Ar, H <sub>2</sub> - n - C <sub>4</sub> H <sub>10</sub> , H <sub>2</sub> - SF <sub>6</sub> , He - Ar

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Author(s)	Reference	System(s)
Suetin, P. E., G. T. Scheholev and R. A. Klestov	Zh. Techn. Fiz. <u>29</u> , 1058 (1959) Trans., Sov. Phys.- Tech. Phys. <u>4</u> , 964 (1960)	H <sub>2</sub> - He, H <sub>2</sub> - AIR, H <sub>2</sub> - CO <sub>2</sub> , He - AIR, He - CO <sub>2</sub>
Suetin, P. E. and B. A. Ivakin	Zh. Techn. Fiz. <u>31</u> , 499 (1961) Trans., Sov. Phys.- Tech. <u>6</u> , 359 (1961)	H <sub>2</sub> - He, H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - AIR, H <sub>2</sub> - Ar, H <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - SF <sub>6</sub> , He - C <sub>2</sub> H <sub>2</sub> , He - N <sub>2</sub> , He - AIR, He - O <sub>2</sub> , He - Ar, He - CO <sub>2</sub> , He - SF <sub>6</sub> , N <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - SF <sub>6</sub> , AIR - C <sub>2</sub> H <sub>2</sub> , AIR - CO <sub>2</sub> , AIR - SF <sub>6</sub> , O <sub>2</sub> - C <sub>2</sub> H <sub>2</sub> , O <sub>2</sub> - CO <sub>2</sub> , O <sub>2</sub> - SF <sub>6</sub> , Ar - C <sub>2</sub> H <sub>2</sub> , Ar - CO <sub>2</sub> , Ar - SF <sub>6</sub> , CO <sub>2</sub> - SF <sub>6</sub>
Suetin, P. E.	<u>Heat and Mass Transfer</u> Vol. 1, Thermophysical Characteristics of Materials and Methods for their Determination, p. 188- 190, (1962) Trans. ORNL Tr-316 (1964)	H <sub>2</sub> - He, H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - AIR, H <sub>2</sub> - Ar, H <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - SF <sub>6</sub> , He - C <sub>2</sub> H <sub>2</sub> , He - N <sub>2</sub> , He - AIR, He - O <sub>2</sub> , He - Ar, He - CO <sub>2</sub> , He - SF <sub>6</sub> , N <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - SF <sub>6</sub> , AIR - CO <sub>2</sub> , AIR - C <sub>2</sub> H <sub>2</sub> , AIR - SF <sub>6</sub> , O <sub>2</sub> - C <sub>2</sub> H <sub>2</sub> , O <sub>2</sub> - CO <sub>2</sub> , O <sub>2</sub> - SF <sub>6</sub> , Ar - C <sub>2</sub> H <sub>2</sub> , Ar - CO <sub>2</sub> , Ar - SF <sub>6</sub> , CO <sub>2</sub> - SF <sub>6</sub>
Summerhays, W. E.	Proc. Phys. Soc. (London) <u>42</u> , 218 (1930)	H <sub>2</sub> O - AIR

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Author (s)	Reference	System(s)
Timmerhaus, K. D. and H. G. Drickamer	J. Chem. Phys. <u>19</u> , 1242 (1951)	CO <sub>2</sub> - CO <sub>2</sub>
Toepler, M.	Ann. Physik <u>58</u> , 599 (1896) (In German)	NH <sub>3</sub> - AIR, CO <sub>2</sub> - AIR
Topley, B. and R. Whytlaw-Gray	Phil. Mag. <u>4</u> , 873 (1927)	AIR - I <sub>2</sub>
Trautz, M. and O. Ludwig	Ann. Physik <u>5</u> , 887 (1930) (In German)	H <sub>2</sub> - BENZENE, O <sub>2</sub> - BENZENE
Trautz, M. and W. Ries	Ann. Physik <u>8</u> , 163 (1931) (In German)	H <sub>2</sub> - BENZENE, H <sub>2</sub> - CCl <sub>4</sub>
Trautz, M. and W. Muller	Ann. Physik <u>22</u> , 353 (1935)	H <sub>2</sub> - ACETONE, O <sub>2</sub> - CCl <sub>4</sub>
Trautz, M. and W. Muller	Ann. Physik <u>22</u> , 333 (1935)	H <sub>2</sub> O - H <sub>2</sub> , H <sub>2</sub> O - AIR, H <sub>2</sub> O - CO <sub>2</sub> , ETHANOL - H <sub>2</sub> , ETHANOL - AIR, ETHANOL - CO <sub>2</sub> , ETHYL ETHER - H <sub>2</sub> , ETHYL ETHER - AIR, ETHYL ETHER - CO <sub>2</sub> , BENZENE - H <sub>2</sub> , BENZENE - O <sub>2</sub> , I <sub>2</sub> - AIR, I <sub>2</sub> - N <sub>2</sub> , Hg - N <sub>2</sub> , Hg - AIR
Vaillant, M. P.	J. Phys. <u>1</u> , 887 (1911) (In French)	AIR - CH <sub>3</sub> OH, AIR - C <sub>3</sub> H <sub>7</sub> OH

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author(s)	Reference	System(s)
Visner, S.	Atomic Energy Commission Report K-688 (Carbide and Carbon Chemicals Company), 1951	Xe - Xe
Visner, S.	Phys. Rev. <u>82</u> , 297 (1951)	Xe - Xe
Vyshenskaya, V. F. and N. D. Kosov	Issledovanie Protessov Perenosa Voprosy Teorii Otnositel Nosti, 114 (1959)	H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub>
Vyshenskaya, V. F. and N. D. Kosov	<u>Heat and Mass Transfer</u> , First All Union Conference, Minsr, June 5-9, 1961, pp. 181-7 Translated by A. L. Monks, Ornl-Tr-506	H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - I <sub>2</sub> , CO <sub>2</sub> - I <sub>2</sub>
Wagner, C.	Z. Physik. Chem (Leipzig) <u>A192</u> , 85 (1943) (In German)	H <sub>2</sub> O - AIR
Waitz, K.	Ann. Physik <u>17</u> , 201 (1882) (In German)	AIR - CO <sub>2</sub>
Waitz, K.	Ann. Physik <u>17</u> 351 (1882) (In German)	AIR - CO <sub>2</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System(s)
Waldmann, L.	Naturwiss. <u>32</u> , 222 (1944)	H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - Ar, H <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - D <sub>2</sub> , N <sub>2</sub> - O <sub>2</sub> , N <sub>2</sub> - Ar, N <sub>2</sub> - CO <sub>2</sub> , O <sub>2</sub> - Ar, O <sub>2</sub> - CO <sub>2</sub> , Ar - CO <sub>2</sub>
Walker, R. E. and A. A. Westenberg	J. Chem. Phys. <u>29</u> , 1139 (1958)	He - N <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub>
Walker, R. E. and A. A. Westenberg	J. Chem. Phys. <u>29</u> , 1147 (1958)	He - N <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub>
Walker, R. E.	J. Chem. Phys. <u>34</u> , 2196 (1961)	O - O <sub>2</sub>
Walker, R. E. and A. A. Westenberg	J. Chem. Phys. <u>31</u> , 519 (1959)	He - Ar
Walker, R. E. and A. A. Westenberg	Personal Communication	He - Ar, He - O <sub>2</sub> , O <sub>2</sub> - CO <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> O - O <sub>2</sub> , CH <sub>4</sub> - O <sub>2</sub> , CO - O <sub>2</sub> , He - N <sub>2</sub>
Walker, R. E. and A. A. Westenberg	J. Chem. Phys. <u>32</u> , 436 (1960)	H <sub>2</sub> - O <sub>2</sub> , CH <sub>4</sub> - O <sub>2</sub> , H <sub>2</sub> O - O <sub>2</sub> , CO - O <sub>2</sub> , O <sub>2</sub> - CO <sub>2</sub>
Walker, R. E. and A. A. Westenberg	Personal Communication	H <sub>2</sub> - O <sub>2</sub> , CH <sub>4</sub> - O <sub>2</sub> , CO - O <sub>2</sub> , O <sub>2</sub> - CO <sub>2</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author(s)	Reference	System(s)
Walker, R. E., N. deHaas and A. A. Westerberg	J. Chem. Phys. <u>32</u> , 1314 (1960)	He - CO <sub>2</sub> , N <sub>2</sub> - CO <sub>2</sub>
Wall, F. T. and G. A. Kidder	J. Phys. Chem. <u>50</u> , 235 (1946)	CO <sub>2</sub> - N <sub>2</sub> O, CO <sub>2</sub> - C <sub>3</sub> H <sub>8</sub> , N <sub>2</sub> O - C <sub>3</sub> H <sub>8</sub> , CO <sub>2</sub> - C <sub>2</sub> H <sub>4</sub> O, N <sub>2</sub> O - C <sub>2</sub> H <sub>4</sub> O
Wasik, S.	J. Chem. Phys. <u>44</u> , to be published (1966)	H <sub>2</sub> - O <sub>2</sub> , He - Ne, He - N <sub>2</sub> , He - O <sub>2</sub> , He - Ar, He - Kr
Watts, H.	Can. J. Chem. <u>43</u> , 431 (1965)	He - Xe, Ne - Xe, Ar - Xe, Kr - Xe, Xe - Xe
Watts, H.	Trans. Faraday Soc. <u>60</u> , 1745 (1964)	He - Kr, Ne - Kr, Ar - Kr, Kr - Kr, Kr - Xe
Weissman, S. and E. A. Mason	J. Chem. Phys. <u>36</u> , 794 (1962)	H - H, H - H <sub>2</sub>
Weissman, S., S. C. Saxena and E. A. Mason	Phys. Fluids <u>4</u> , 643 (1961)	Ne - CO <sub>2</sub>
Weissman, S., and E. A. Mason	J. Chem. Phys. <u>37</u> , 1289 (1962)	H <sub>2</sub> - HD, H <sub>2</sub> - He, H <sub>2</sub> - D <sub>2</sub> , H <sub>2</sub> - Ne, H <sub>2</sub> - H <sub>2</sub> , H <sub>2</sub> - CH <sub>4</sub> , H <sub>2</sub> - N <sub>2</sub> , H <sub>2</sub> - CO, H <sub>2</sub> - O <sub>2</sub> , H <sub>2</sub> - Ar, H <sub>2</sub> - Xe, HD - D <sub>2</sub> , He <sup>3</sup> - He <sup>4</sup> , D <sub>2</sub> - Ne, He - Ne, Ne - Ar, He - Kr, He - Xe, Ne - Ne, Ne - Ar, Ne - Kr,

## BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Weissman, S. and E. A. Mason - Cont.		Ne - Xe, N <sub>2</sub> - N <sub>2</sub> , N <sub>2</sub> - CO, N <sub>2</sub> - O <sub>2</sub> , CO - O <sub>2</sub> , O <sub>2</sub> - O <sub>2</sub> , Ar - Ar, Ar - Kr, Ar - Xe, Kr - Xe
Weissman, S.	J. Chem. Phys. <u>40</u> , 3397 (1964)	HCl - HCl, HBr - HBr, BF <sub>3</sub> - BF <sub>3</sub> , UF <sub>6</sub> - UF <sub>6</sub> , CH <sub>4</sub> - CH <sub>4</sub> , CO - CO, CO <sub>2</sub> - CO <sub>2</sub> , H <sub>2</sub> - NH <sub>3</sub> , N <sub>2</sub> - NH <sub>3</sub> , O <sub>2</sub> - NH <sub>3</sub> , CO <sub>2</sub> - HCl, NH <sub>3</sub> - CH <sub>4</sub> , SO <sub>2</sub> - CO <sub>2</sub> , (C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub> O - H <sub>2</sub> , C <sub>2</sub> H <sub>4</sub> - NH <sub>3</sub> , HCl - H <sub>2</sub> , SO <sub>2</sub> - H <sub>2</sub> , CCl <sub>4</sub> - CH <sub>2</sub> Cl <sub>2</sub> , CCl <sub>4</sub> - CHCl <sub>3</sub> , CHCl <sub>3</sub> - (C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub> O, N <sub>2</sub> O - CO <sub>2</sub> , CO <sub>2</sub> - H <sub>2</sub> , N <sub>2</sub> O - H <sub>2</sub> , NO - H <sub>2</sub> , CO <sub>2</sub> - N <sub>2</sub> , N <sub>2</sub> O - NO, NO - N <sub>2</sub> , Ar - C <sub>2</sub> H <sub>4</sub> , C <sub>3</sub> H <sub>6</sub> - He, 2 - C <sub>4</sub> H <sub>8</sub> - He, C <sub>2</sub> H <sub>2</sub> - H <sub>2</sub> , C <sub>2</sub> H <sub>4</sub> - H <sub>2</sub> , C <sub>2</sub> H <sub>6</sub> - H <sub>2</sub> , C <sub>3</sub> H <sub>6</sub> - H <sub>2</sub> , C <sub>3</sub> H <sub>8</sub> - H <sub>2</sub> , 2 - C <sub>4</sub> F <sub>8</sub> - H <sub>2</sub> , C <sub>2</sub> H <sub>4</sub> - N <sub>2</sub> , O <sub>2</sub> - C <sub>2</sub> H <sub>4</sub> , C <sub>2</sub> H <sub>4</sub> - CO, CO <sub>2</sub> - CH <sub>4</sub> , C <sub>3</sub> H <sub>8</sub> - N <sub>2</sub> O, C <sub>3</sub> H <sub>8</sub> - CO <sub>2</sub> , C <sub>2</sub> H <sub>2</sub> - CH <sub>4</sub> , C <sub>2</sub> H <sub>6</sub> - CH <sub>4</sub> , C <sub>3</sub> H <sub>8</sub> - CH <sub>4</sub> , C <sub>3</sub> H <sub>6</sub> - C <sub>2</sub> H <sub>2</sub> , C <sub>7</sub> F <sub>16</sub> - C <sub>8</sub> H <sub>18</sub> , C <sub>3</sub> H <sub>8</sub> - C <sub>2</sub> H <sub>6</sub> , C <sub>3</sub> H <sub>8</sub> - C <sub>3</sub> H <sub>6</sub> , CCl <sub>4</sub> - C <sub>6</sub> H <sub>6</sub>



BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Weissman, S.	<u>Advances in Thermophysical Properties at Extreme Temperatures and Pressure</u> , ASME, New York, 1965 pp. 12-18	He - Ne, He - Ar, He - Kr, He - Xe, Ne - Ar, Ne - Kr, Ne - Xe, Ar - Kr, Ar - Xe, Kr - Xe
Wendt, R. P., J. N. Mundy, S. Weissman and E. A. Mason	Phys. Fluids <u>6</u> , 572 (1963)	CO <sub>2</sub> - CO <sub>2</sub> , Kr - Kr
Westenberg, A. A. and R. E. Walker	J. Chem. Phys. <u>26</u> , 1753 (1957)	He - N <sub>2</sub> , N <sub>2</sub> - Ar, N <sub>2</sub> - CO <sub>2</sub>
Westenberg, A. A. and G. Frazier	J. Chem. Phys. <u>36</u> , 3499 (1962)	H <sub>2</sub> - Ar
Winkelmann, A.	Ann. Physik <u>22</u> , 1 (1884) (In German)	H <sub>2</sub> - C <sub>2</sub> H <sub>5</sub> OH, AIR - C <sub>2</sub> H <sub>5</sub> OH, CO <sub>2</sub> - C <sub>2</sub> H <sub>5</sub> OH, H <sub>2</sub> - ETHER, CO <sub>2</sub> - ETHER, H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O - AIR, H <sub>2</sub> O - CO <sub>2</sub>
Winkelmann, A.	Ann. Physik <u>22</u> , 152, (1884) (In German)	H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O - AIR, H <sub>2</sub> O - CO <sub>2</sub>
Winkelmann, A.	Ann. Physik <u>23</u> , 203 (1884) (In German)	H <sub>2</sub> , AIR AND CO <sub>2</sub> (EACH WITH THE FOLLOWING ESTERS) - ETHYL FORMATE, PROPYL FORMATE, METHYL ACETATE, ETHYL ACETATE, iso - BUTYL ACETATE, METHYL PROPIONATE,

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Winkelmann, A. - Cont.		ETHYL PROPIONATE, PROPYL PROPIONATE, AMYL PROPIONATE, METHYL BUTYRATE, ETHYL BUTYRATE, PROPYL BUTYRATE, iso - BUTYL BUTYRATE, METHYL iso - BUTYRATE, ETHYL iso - BUTYRATE, PROPYL iso - BUTYRATE, iso - BUTYL iso - BUTYRATE, AMYL iso - BUTYRATE, ETHYL VALEREATE, PROPYL VALEREATE, iso - BUTYL VALEREATE,
Winkelmann, A.	Ann. Physik 26, 105 (1885) (In German)	H <sub>2</sub> , AIR AND CO <sub>2</sub> (EACH WITH THE FOLLOWING COMPOUNDS) - FORMIC ACID, ACETIC ACID, PROPIONIC ACID, iso - BUTYRIC ACID, n - BUTYRIC ACID, iso - VALERIC ACID, METHYL ALCOHOL, ETHYL ALCOHOL, PROPYL ALCOHOL, iso - BUTYL ALCOHOL, n - BUTYL ALCOHOL, AMYL ALCOHOL, n - AMYL ALCOHOL, n - HEXYL ALCOHOL

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Winkelmann, A.	Ann. Physik, <u>33</u> , 445 (1888) (In German)	H <sub>2</sub> O - AIR
Winkelmann, A.	Ann. Physik <u>36</u> , 93 (1889) (In German)	H <sub>2</sub> - H <sub>2</sub> O, H <sub>2</sub> O - AIR, H <sub>2</sub> O - CO <sub>2</sub>
Winn, E. B. and E. P. Ney	Phys. Rev <u>72</u> , 77 (1947)	CH <sub>4</sub> - CH <sub>4</sub>
Winn, E. B.	Phys. Rev. <u>74</u> , 698 (1948)	N <sub>2</sub> - N <sub>2</sub>
Winn, E. B.	Phys. Rev. <u>80</u> , 1024 (1950)	CH <sub>4</sub> - CH <sub>4</sub> , Ne - Ne, N <sub>2</sub> - N <sub>2</sub> , O <sub>2</sub> - O <sub>2</sub> , Ar - Ar, CO <sub>2</sub> - CO <sub>2</sub>
Winter, E. R. S.	Trans. Faraday Soc. <u>47</u> , 342 (1951)	N <sub>2</sub> - N <sub>2</sub> , O <sub>2</sub> - O <sub>2</sub> , CO <sub>2</sub> - CO <sub>2</sub>
Wintergerst, E.	Ann. Physik <u>4</u> , 323 (1930) (In German)	NH <sub>3</sub> - AIR
Wise, H.	J. Chem. Phys. <u>31</u> , 1414 (1959)	H - H <sub>2</sub>
Wise, H.	J. Chem. Phys. <u>34</u> , 2139 (1961)	H - H <sub>2</sub>
Wretschko, A.	Sitzber., Akad, Wiss., Wien <u>62</u> , 575 (1870) (In German)	H <sub>2</sub> - CO <sub>2</sub> , O <sub>2</sub> - CO <sub>2</sub>

BIBLIOGRAPHY OF DIFFUSION DATA FOR DILUTE BINARY GAS MIXTURES - Cont.

Author (s)	Reference	System (s)
Young, R. A.	J. Chem. Phys. <u>34</u> , 1295 (1961)	N - N <sub>2</sub>

#### IV LIQUIDS

Data on the liquid phase molecular transport coefficients is being compiled and preliminary evaluation of some systems has begun. Both aqueous and non-aqueous solvent systems with ionizing solutes are included in the program. Excluded initially from this compilation and evaluation survey are those systems in which the solvent or the solute represents a heterogeneous chemical species such as some of the liquid petroleum mixtures.

In compiling and evaluating viscosity and diffusivity data close consideration is given to the procedures of experimentation and analysis, and the physical equipment used by each investigator. When possible, existing chemical-physical theories of viscosity and diffusivity in gases, liquid and solids will be used as a guide for data evaluation and correlation.

Data on the liquid phase molecular transport coefficients is being compiled by Dr. Beckmann. Compilation and evaluation of certain systems has begun. Both aqueous and non-aqueous solvent systems with ionizing solutes are included in the program. Excluded initially from this compilation and evaluation survey are those systems in which the solvent or the solute represents a heterogeneous chemical species that can not be readily characterized on the basis of pure component properties--for example, a liquid tar or broad spectrum liquid petroleum of generally variable composition and properties would not be included, but a binary, ternary or multicomponent mixture (either solvent or solute) of pure components with a definable composition is to be included.

A convenient format for the initial characterization of liquid systems has been adopted:

- I. Polar Solvents with Ionizing or Non-Ionizing Solutes.
- II. Non-Polar Organic Solvents with Ionizing or Non-Ionizing Solutes.

### III. Inorganic and Liquid Metal Solvents with Organic or Inorganic Solutes.

The literature has been surveyed to permit a general classification and delineation of the experimental techniques and the theories of liquid phase molecular transport have been reviewed and collated to enable the definition of the various possible transport coefficients---integral, differential, etc.

Because of the large scope of each of the above areas, the initial phases of the literature survey have been directed primarily at the last five years (1960-1965). One of the reasons for this initial limitation was that this period would afford a barometer for the sources of current information and serve as a guide for continuing efforts; also, the current literature is least likely to be available in compendiums available to researchers in the field. Some of the more significant earlier collections have also been included in these initial compilations.

#### EXPERIMENTAL METHODS

Technique for measuring diffusion coefficients in liquids are in principle the same as those employed with gases. Diffusion coefficients in liquid are smaller than those in gases by at least  $10^4$ . Differences in the experimental set up, to be used with liquids, compared with that employed with gases, are largely due to this fact.

A certain concentration distribution, obtained with a given apparatus, is essentially a function of the Fourier number,  $X^2/Dt$ , if X refers to the linear dimensions of the apparatus, and if D and t are diffusion coefficient and time, respectively. With linear dimensions kept constant a decrease in diffusion coefficient by  $10^4$  would involve an increase in time of observation by the

same factor. Hence, if we are interested in not extending the time of observation beyond that necessary for gases we are forced to use diffusion apparatus of only  $10^{-2}$  the size of those employed with gases. Thus we arrive at linear dimensions of the order of magnitude of 1 cm., or in some special arrangements, even less. Although it is possible to work with diffusion cells of about this size, we may be compelled by the method of analysis available to increase the dimensions of the cell, and consequently to deal with much longer times of observation. If normal chemical analysis is chosen, as has been the case especially in the earlier work on diffusion in liquids, it may be necessary to increase the linear dimensions by as much as a factor of 10, thus increasing the time of observation by a factor of 100. If, on the other hand, appropriate physical methods of analysis are available, dimensions of 1 cm may be sufficient.

A large number of techniques have been used to measure liquid diffusivities. The following Table summarizes some of these. The stationary methods and the non-stationary closed system methods are very similar to the methods described above for gases. They differ from the gas techniques primarily in the method of analysis of the concentration. Optical techniques for measuring concentration have been used much more widely in liquids than in gases.

#### PRELIMINARY FINDINGS

The critical evaluation of liquid diffusivity data is complicated by the fact that no satisfactory theory of the liquid state is presently available. Most of the available theories on diffusion lack the generality required for their application to a wide variety of systems.

Table I

MEASUREMENT OF LIQUID DIFFUSIVITIES

I. Stationary and Quasi-stationary

- A. Long Vessels (tubular or rectangular connecting two containers)
- B. Diaphragm Cell-Horizontal sintered glass disks between an upper and a lower cell
  - 1. Northrup-Anson Cell-higher concentration on top; no stirring
  - 2. Stokes Cell-higher concentration below; magnetically stirred in both sections

II. Non-Stationary

- A. Open System-Fritted porcelain filled with solute to equilibration. Then allowed to diffuse to a large constant concentration volume of solution.
  - 1. Diffusion rate measured by loss in wt. of fritted porcelain
  - 2. Diffusion rate measured by tracer counting
- B. Closed System-Diffusion is restricted within the cell
  - 1. Restricted Diffusion
    - a. Layer analysis- 4 to 6 layers of solution are analyzed using tables prepared for such treatment
    - b. Conductometric-changes in the conductance at different points are measured
    - c. Absorption-Gold and mercury hydrosols are used to detect change in concentration
    - d. Colorimetric-constant concentration is followed in the cell by changing the observation point
    - e. Total reflection-concentration is followed as in (d) by the use of prisms
    - f. Sonic technique-measure change in velocity of sound
  - 2. Free Diffusion-Optical
    - a. Refractive index gradient is measured
    - b. Refractive index is measured



The available correlations for the prediction of diffusion coefficients may be divided into two classes. In the first class, theoretical correlations based on the works of Nernst, Einstein, Eyring and Hill are included. The diffusivities predicted by these correlations show high deviation from experimentally determined diffusivities which renders them of little use for engineering design purposes. The second class of correlation includes the empirical equations of Wilke and Chang, Othmar and Thakar, Kamal and Canjar. Although these equations are fundamentally empirical, they are based on the results of the theoretical considerations. At the present time, the best available correlation for the calculation of diffusion coefficients is the empirical correlation of Wilke and Chang. Their equation gives values of the diffusion coefficient with an average deviation of 11% from the experimental values. (Kamal, M. R. and L. N. Canjar, A.I.Ch. E. Journal, 8, 329 (1962).)

In most of the available correlations, viscosity of the system under consideration is one of the controlling variables. This makes it necessary to have available viscosity data on the system for the calculation of the diffusion coefficient. Accordingly, any error that may occur in the determination of the viscosity will add to the uncertainty of the calculated diffusion coefficient. Besides, very little data is available on the viscosity of solutions which makes it difficult to study the effect of concentration on the diffusion coefficient.

The results obtained for 56 different liquid-liquid systems with eight different solvents are reported in Table II. The deviations of the calculated diffusivities based on Wilke's and Kamal's equations are also reported.

It is possible to reduce the error in the results by noting that systems with acetic acid as a solute show high negative

deviation. This may be explained by the fact that in the calculation of the latent heat term, the molecular weight was assumed to be 100. The formula weight of acetic acid is 60.07, but usually a molecular weight of 100 is assumed in the calculation of the molar heat of vaporization due to association on boiling. The amount of error may be reduced more than 1% by basing the calculation on a molecular weight of 80 rather than 100. Similar arguments can be made to improve the results on formic acid and other associating compounds.

It was not possible to obtain any correlation between the heat of solution at infinite dilution and the deviation from the experimental values.

An analysis of the variation of diffusion coefficient with temperature was made. Although not complete due to the lack of sufficient data, it was apparent that for the system acetic acid-carbon tetrachloride, the temperature coefficient of the diffusivity obtained from Wilke's equation was higher than the experimental temperature coefficient.

The binary diffusion data in water for 47 compounds and elements are presented in Table III. These data illustrate a serious problem in the critical evaluation of liquid diffusion data. Namely, that only in a few instances has a system been studied by more than one investigator. The gases  $N_2$ ,  $O_2$ ,  $H_2$ , and He are good examples of substances where several investigators have measured the diffusivity. In these instances the agreement between investigators is of the same order as the deviations from existing correlations.

Table III also presents the diffusivity values predicted for the system using three of the existing correlations. For these aqueous diffusivities the difference has been measured and calculated averages about 15% which is somewhat higher than the

results shown in Table II for all types of solvents.

The diffusivity of amines in benzene is presented in Table IV. The agreement between experiment and the various correlations is somewhat better than shown in Table II and III.

Experimental data in liquid diffusion from 147 literature reference has been obtained and is on file. Some of the data are presented in Tables II, III and IV.

Table II

## DIFFUSION COEFFICIENTS AT INFINITE DILUTION

Solvent	Solute	Temp. °C.	$10^5 \times$ D <sub>exp.</sub> cm <sup>2</sup> /sec	Kamal's Percent Deviation	Wilke's Percent Deviation
Benzene	Acetic Acid	15	1.92	-29	+17
	Benzaldehyde	15	1.73	-19	-11
	Benzene	25	2.14	-6	
	Bromobenzene	15	1.86	-24	-17
	Carbon tetrachloride	25	1.91	+9	+6
	Chlorobenzene	15	1.42	+8	+11
	Chloroform	15	2.11	-7	-10
	Ethyl ether	15	2.21	+3	+1
	Ethylene bromide	15	1.97	-13	-13
	Formic acid	6	1.99	+26	+22
	Isoamyl alcohol	15	1.48	-9	-1
	Methanol	15	2.00	-15	
	Nitrobenzene	15	1.84	-25	-19
	Phenol	5	1.27	-10	+8
	Propanol	15	1.60	-17	+20
Carbon Tetrachloride	Acetic acid	25	1.50	-31	+6
	Benzene	25	1.38	+6	
	Carbon tetrachloride	25	1.41	+8	+23
	Formic acid	25	1.89	+4	+6
Chlorobenzene	Bromobenzene	25	1.64	-11	-13
	Toluene	25	1.69	+2	-5

Table II (Continued)

Solvent	Solute	Temp. °C.	$10^5 \times$ Disp. cm/sec	Kamal's Percent Deviation	Wilke's Percent Deviation
Bromobenzene	Chlorobenzene	25	1.26	+ 9	- 7
	Toluene	25	1.41	+10	
Toluene	Acetic acid	25	2.26	-34	-11
	Bromobenzene	25	2.27	-31	
	Carbon tetrachloride	25	2.19	0	+ 1
	Chlorobenzene	25	2.21	-26	
	Formic acid	25	2.64	+ 7	- 4
Ethanol	Methyl iodide	7.5	2.23	+ 7	+ 7
	Isoamyl alcohol	20	0.87	+24	+ 9
	Pyridine	20	1.12	- 8	-11
Methanol	Water	25	1.13	-12	
	Acetone	15	2.50	- 4	
	Allyl alcohol	15	1.80	-13	+13
	Aniline	15	1.49	- 1	+ 8
	Benzaldehyde	15	1.66	- 4	- 7
	Bromobenzene	15	1.75	- 7	-12
	Carbon tetrachloride	15	1.70	+36	- 1
	Chloral hydrate	15	1.16	+18	+32
	Chloroform	15	2.07	+17	- 9
	Ethyl bromide	15	2.40	+13	-16
	Ethylene bromide	15	1.95	0	-12
	Furfural	15	1.70	-13	+ 3

Table II (Continued)

Solvent	Solute	Temp. °C.	$10^5 \times$ Disp. cm <sup>2</sup> /sec	Kamal's Percent Deviation	Wilke's Percent Deviation
Water	Isoamyl alcohol	15	1.34	+15	+ 7
	Phenol	15	1.40	- 6	+15
	Propionic acid	15	1.62	+25	+12
	Pyridine	15	1.58	+14	+11
	Water	15	1.78	- 7	
	Acetic acid	15	0.91	+ 3	+ 9
	Acetone	15	1.25	+15	
	Allyl alcohol	15	0.90	+ 4	+ 8
	n-Butyl alcohol	15	0.77	+ 8	+ 3
	Ethanol	15	1.00	- 1	+11
	Methanol	15	1.28	- 9	+ 8
	Propanol	15	0.87	+ 4	+ 5
	Isoamyl alcohol	15	0.69	+33	+ 1

Table III

BINARY DIFFUSION COEFFICIENTS IN WATER AT INFINITE DILUTION ( $\text{cm}^2 \text{sec}^{-1} \times 10^5$ )

Solute	T(°C)	DATA		CALCULATED			% DEVIATION			Othmar-- Thakar
		Exper.	Ref.	Wilke	Scheibel	Thakar	Wilke	Scheibel		
Acetamide	25	1.261	26	1.68	1.67	1.58	33.2	32.4	25.3	
Acetanilide	25	0.820	22	0.869	0.844	0.814	5.98	2.9	-.73	
B-Alanine	25	0.9419	35	1.09	1.022	1.02	15.72	7.83	7.82	
2-Amino-n-Butyric Acid	25	0.8305	40	0.975	0.905	0.905	17.4	8.97	8.97	
Argon	25	2.02	10	7.00	4.34	6.68	246	114.85	230	
Argon		1.92	11				264	126.04	247.91	
Benzene	25	1.14	108	1.062	1.015	1.015	-6.84	-10.96	-10.96	
Benzene		1.10	140				-3.454	-7.72	-7.72	
Benzene		1.08	140				-1.66	-6.02	-6.02	
Benzoic Acid	25	0.935	127	0.944	0.912	0.905	.96	-2.45	-3.2	
Butanol	25	0.851	51	0.79	0.73	0.75	-7.168	-14.22	-11.87	
Bovine Mer-Captalbumium	25	0.0589	132							
C <sub>2</sub> H <sub>6</sub>	25	1.88	11	1.555	1.61	1.488	-17.28	-14.36	-20.85	
C <sub>2</sub> H <sub>6</sub>		1.71	10				-9.06	-5.8	-12.98	
C <sub>2</sub> H <sub>4</sub>	25	2.00	11	1.609	1.65	1.54	-19.55	-17.5	-23.0	
C <sub>2</sub> H <sub>4</sub>		1.59	10				1.2	3.6	-3.1	
CO <sub>2</sub>	18	1.79	138	1.633	1.62	1.54	-8.77	-9.49	-13.96	
CO <sub>2</sub>		1.81	136				-9.77	-10.49	-14.91	
CO <sub>2</sub>		1.83	120				-10.92	-11.47	-15.84	
Dextrose	25	0.6765	88	0.755	0.79	0.706	11.6	16.78	4.36	
Erythritol	25	0.805	87	0.942	0.873	0.88	17.01	8.45	9.31	
Ethanol	25		56	1.20	1.41	1.33				
Ethyl Acetate	25	0.909	54	1.062	1.015	1.015	16.8	11.6	11.6	

Table III (Continued)

Solute	T(°C)	DATA			CALCULATED			% DEVIATION		
		Exper.	Ref.	Wilke	Scheibel	Othmar-Thakar	Wilke	Scheibel	Othmar-Thakar	
Ethylene Glycol	25	1.153	88	1.234	1.19	1.035	7.02	3.2	10.23	
Formimide	25	1.6091	3	1.76	1.77	1.682	9.38	10	4.53	
Glycerin	25	1.0609	40	1.09	1.025	1.021	2.74	-3.38	-3.76	
Glycolamide	25	1.142	87	1.285	1.215	1.2	12.52	6.39	5.08	
Glycine	25	1.0595	40	1.234	1.19	1.035	16.4	12.3	2.31	
Glycylglycine	25	0.7963	40	0.853	0.825	0.818	6.6	3.6	2.65	
He	25	4.8	47	2.53	2.74	2.42	-47.3	-42.91	-49.58	
He		6.3	107				-59.84	-56.50	-61.59	
He		9.5	10				-73.36	-71.15	-74.52	
He		8.02	88				-68.45	-65.84	-69.82	
iso-Butanol	25	0.983	54	0.79	0.73	0.75	-19.63	-25.74	-23.70	
Lactimide	25	0.993	144	0.675	0.58	0.635	-32.02	-41.5	-36.05	
Methyl Vrea	25	1.168	106	1.234	1.19	1.035	5.65	1.88	11.38	
Di-Methyl Vrea	25	0.998	106	1.062	1.015	1.015	6.41	1.7	1.7	
N <sub>2</sub>		2.14	22	1.97	2.00	1.86	-7.94	-6.54	-13.084	
N <sub>2</sub>		1.75	22				12.57	14.28	6.28	
B-Naphthal	25	1.14	86	0.832	0.77	0.78	-27.01	-32.46	-31.58	
O <sub>2</sub>	25	2.41	107	2.29	2.49	2.26	-4.97	3.32	6.22	
O <sub>2</sub>		2.55	22				-10.19	-2.35	-11.37	
O <sub>2</sub>		2.03	22				12.80	22.66	11.33	
O <sub>2</sub>		3.78	63				-39.42	-34.12	-40.21	
Pentaerythritol	20	0.732	146	0.74	0.70	0.69	1.1	-4.37	-5.74	
Propionamide	25	1.093	87	1.118	1.12	1.11	2.28	2.47	1.55	
Propylene	25	1.44	138	1.295	1.25	1.235	-10.07	-13.2	-14.25	
Raffinose	25	0.441	38	0.372	0.398	0.347	-15.6	-9.75	-21.31	
Sucrose	25	0.552	74	0.512	0.51	0.48	-7.24	-7.25	-13.04	
Toluene	25	0.955	18	0.944	0.912	0.904	-1.15	-4.5	-5.34	
Toluene		0.975	18				-3.18	-6.46	-7.28	



Table III (Continued)

Solute	T(°C)	<u>DATA</u>		<u>CALCULATED</u>				<u>% DEVIATION</u>			
		Exper.	Ref.	Wilke	Scheibel	Othmar-Thakar	Wilke	Scheibel	Othmar-Thakar	Scheibel	Othmar-Thakar
Thio-Vrea	25	1.331	106	1.295	1.25	1.235	-2.7	-6.01	-7.21		
Vrea	25	1.382	106	1.485	1.465	1.42	7.45	6	2.75		
dl-Valine	25	0.7722	49	0.87	0.842	0.814	12.66	8.29	5.14		
D <sub>2</sub> O	25	2.2605	88	2.78	3.02	2.7	22.9	33.6	19.44		
H <sub>2</sub>	25	4.1	55	2.53	2.74	2.42	-16.58	-3.17	-21.95		
H <sub>2</sub>		4.27	22				-19.9	-7.02	-25.05		
H <sub>2</sub>		4.8	107				-28.75	-17.29	-33.33		
H <sub>2</sub>		5.3	22				-35.47	-25.09	-39.62		
H <sub>2</sub>		5.26	22				-34.98	-24.52	-39.16		
Glycerol	25	0.938	106	1.09	1.025	1.021	16.2	8.49	8.13		

Table IV

DIFFUSIVITY OF AMINES IN BENZENE AT 15°C

	<u>Wilke Correlation</u>		<u>Scheibel</u>		<u>Othmer-Thakar</u>		<u>Experimental</u>
	value $\frac{\text{cm}^2}{\text{sec}}$	%error	value $\frac{\text{cm}^2}{\text{sec}}$	%error	value $\frac{\text{cm}^2}{\text{sec}}$	%error	$\frac{\text{cm}^2}{\text{sec}}$
Bromo-aniline	$1.43 \times 10^{-5}$	+3	$1.53 \times 10^{-5}$	+8	$1.28 \times 10^{-5}$	-9	$1.41 \times 10^{-5}$
Chloro-aniline	$1.46 \times 10^{-5}$	-6	$1.54 \times 10^{-5}$	-1	$1.31 \times 10^{-5}$	-16	$1.56 \times 10^{-5}$
Acetyl-diphenyl-amine	$0.99 \times 10^{-5}$	+11	$1.14 \times 10^{-5}$	+27	$0.89 \times 10^{-5}$	-1	$0.90 \times 10^{-5}$

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## V DIFFUSION IN POLYMER-DILUENT SYSTEMS

### INTRODUCTION

During the past few years there has been a great deal of interest in experimentally determining how small molecules diffuse in polymeric solids due to the increased use of polymeric materials as barriers to flow. A considerable amount of research has been concentrated on the flow of gases through amorphous polymers and semicrystalline polymers. Diffusion through amorphous polymers usually follows Fick's laws while the transfer process through semicrystalline polymers is frequently more complex. The quantity of research on the transfer of liquids through polymeric material is much smaller than that covering gas diffusion and the theoretical aspects of liquid diffusion are not well developed.

The majority of the diffusivity values reported in the literature are obtained by making both solubility or sorption measurements and permeability measurements. However, in recent years there has been a growing tendency to employ the time-lag method of measuring diffusivities developed by Barrer. Although this method requires a rather rapid measuring technique, it has the advantage of requiring only one type of measurement to obtain diffusivity values.

It has been reported in the literature that the diffusion of small molecules through polymeric materials is in general dependent upon temperature, pressure, the structure and size of the diffusing molecules, polymer structure and polymer density. Most of the studies reported in the literature are of a rather limited nature and usually report diffusivities only as a function of temperature.

### DISCUSSION

The work on diffusion in polymer-diluent systems was started in June of this year. The original plan was to evaluate data on the diffusion of gases and water through polyethylene and polystyrene. The major part of the summer was to be spent in collecting these data. As the search progressed during the summer it became apparent that we would be searching the same journals in the future when we began evaluating other systems. To reduce the number of searches, we modified our searching scope so that all systems of interest are now included.

The search to this data has been concentrated in the Journal of Applied Polymer Science, the Journal of Polymer Science, Transactions of the Faraday Society, and the Retrieval Guide to Thermophysical Properties Research Literature edited by Y. S. Touloukian.

We have noted that there is a rather large amount of permeability data and solubility data. However, most experimenters do not make both measurements and therefore the quantity of diffusion results is much smaller than the permeability and solubility results. The first group of references in the bibliography contain diffusion results, while the second group contain permeability and solubility results. The usefulness of the data in the second group in calculating diffusivities is under study.

We have found that there is apparently some rather good data on the diffusion of several gases through polyethylene as a function of polymer density and temperature. This data is being correlated. We have not found a significant amount of data on the diffusion of gases through polystyrene. The polystyrene gas systems may be an area which will require experimental measurements. We are continuing the literature search in this area to determine if this is the case.

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## VI DIFFUSION IN INORGANIC SOLIDS

### INTRODUCTION

This area of investigation was only started in June 1966, because of the transfer of Dr. Homer Schamp from the Institute of Molecular Physics. Since that time the work has been under the direction of Dr. L. Skolnick. The work has been primarily involved with self-diffusion in the silver halides. A fairly extensive overview of this field has been completed and is included in this report. Impurity diffusion in these materials is now being studied. This work was considerably aided by a compilation by Friauf appearing in the 3rd Edition of the "American Institute of Physics Handbook". The literature was surveyed from the date of this compilation through 1965 using Chemical Abstracts and Physics Abstracts as well as the services of the Thermophysical Properties Research Center at Purdue University.

Work was started on the Alkali Halides by surveying the relevant literature cited in the compilation of the "Physics Handbook" and reviewing abstracts from 1960-64 on self-diffusion in Alkali Halides. This work is reported simply as a bibliography and is being continued through the next period of the contract.

The literature citation method used in both the silver halide and alkali halide bibliographies is to use the designation of the "Physics Handbook" for literature cited there. Additional references are indicated by a three digit number following the first letter of the principal author's name. These are numbered chronologically so that further additions may be made without changing designations for the literature.

# DISCUSSION

The data for self-diffusion in the silver halides are presented in Figures 1-7. In general the observed diffusion coefficients are simply plots of the equations presented by Friauf in his "Handbook of Physics" compilation from data from the references cited in the Figures. For consistency we have employed the same numbers for the references as was used in this compilation. To these data we have added more recent results found in the literature, such references are indicated by our own literature citations which are three digit numbers to clarify the fact that they do not occur in the already existing compilation.

In addition to these values of observed diffusivities calculated values of silver diffusivity are also presented in these figures. These are solutions to the Einstein equation:

$$D_{calc} = \frac{\sigma kT}{c} \quad (1)$$

Where the conductivity,  $\sigma$  was obtained from the equations shown in the "Handbook of Physics" compilation, except for  $\beta$  Ag I which will be discussed below, and more recent data where available. Because of the anomolous expansion coefficients of silver halides the concentration of silver ions,  $c$  was carefully evaluated in order to present the  $D_{calc}$  data shown. As will be noted our results do not always agree with others, for the ratio of  $D$  observed to  $D$  calculated from equation (1).

### Silver Chloride

The diffusivity of Ag in Silver Chloride is shown in Fig. 1. Measured values of Ag<sup>110</sup> diffusion are shown from Compton and Maurer (C20) and the more recent data of Reade and Martin (R101). As can be seen the latter data are consistently below the results reported by Compton and Maurer; ranging from about 50% at 400°C to about 20% at 200°C. Reade and Martin suggest an uncertainty of only 10% in their data at these temperatures. The reason for the discrepancy is not clear. Impurity effects are inconsistent since the same authors report anomalously high values of Cl<sup>36</sup> diffusivity as will be discussed below. It seems likely that the sectioning technique used by Compton and Maurer should be considered more reliable than the surface activity measurements of Reade and Martin. On the other hand, the equation suggested by Compton and Maurer and plotted in Figure 1 is incorrect. In comparing the data with the equation:

$$D_{Ag^+}(AgCl) = 1.5 \exp \left( \frac{-0.89}{kT} \right) \quad (2)$$

one finds that the equation gives values consistently above the least squares fit with the data. Therefore, it is suggested that the following equation be adopted:

$$D_{Ag^+}(AgCl) = 3.54 \exp \left( \frac{-0.95}{kT} \right) \quad (3)$$

This equation also agrees much more closely with the data of Reade and Martin.

In comparing the diffusion data with electrical conductivity, the equations presented in the "Physics Handbook" were used,



along with the thermal expansion data of Strelkow (S103). Recalculation using the expansion data of Zeiten (Z101) did not change the results significantly. It is to be noted that the electrical conductivity data of Ebert and Teltow (E3) gives a calculated diffusion coefficient practically identical to equation (3) and in agreement with the data of Compton and Maurer (C-20). The conductivity data also is in fair agreement with some earlier data (T-12, L-1) but does not agree with the equation reported for Compton and Maurer's data as is clear from Figure 1. Our plot was obtained using the equation reported in "Handbook of Physics", but we found no such equation in reference (C-20), since the data presented for  $D_{calc}$  in reference (C-20) is from work performed in 1933 (L-1) which used the equation:

$$\bar{D} = 10^5 \exp \left( -\frac{0.90}{kT} \right) \quad (4)$$

not the reported expression (Physics Handbook).

$$\bar{D} = 2.9 \times 10^4 \exp \left( -\frac{0.82}{kT} \right) \quad (5)$$

In view of the error noted earlier for observed diffusion coefficients we prefer to examine Compton's original data before coming to a final conclusion for the calculated diffusion coefficients. If, however, the data from (E-3) is correct then the interstitialcy mechanism reported by Compton and Maurer (C-20) and Lidiard (L-6) is not substantiated. An interstitial mechanism is suggested by the close agreement of  $D_{obs}$  with  $D_{calc}$  from the Einstein relation.

Figure 2 shows the observed self-diffusion coefficients for chlorine in AgCl. There is good agreement between Compton and Maurer (C-20) and Reade and Martin (R-101) above 350°C.

"Nolting (N103) has substantially lower values for reasons which are not clear. Recently, (Data not shown in Figure 2) Lakatos and Lieser (L107) confirmed the results of (C-20) between 300 and 450°C and showed that the break in the curve reported by Reade and Martin occurred only at 300°C, not 325°C as shown. The two equations reported are:

$$D_{Cl}-(AgCl) = 130 \exp \left( -\frac{1.61}{kT} \right) \quad (5a)$$

$$D_{Cl}-(AgCl) = 85 \exp \left( -\frac{1.58}{kT} \right) \quad (5b)$$

by C-20 and L-107 respectively. This is quite remarkable agreement for such low values of the diffusion coefficient. Since the data of L-107 is valid for a wider range of temperature equation (5b) is recommended. Below 300°C there appears to be an impurity effect requiring further work to clarify the chlorine diffusivity at these low temperatures.

### Silver Bromide

The diffusivity for  $Ag^+$  in AgBr is shown in Figure 3. Diffusion in single crystals was directly observed by Friauf (F-8) and by Miller and Maurer (M-10). These results are in good agreement and therefore the equations presented in the earlier compilation in the "Handbook of Physics" are considered valid. Both (F-8) and (M-10) have measured conductivity. In addition, Teltow (T-2) has measured the conductivity in polycrystalline AgBr. Here again the agreement is quite excellent. In view of Friauf's excellent analysis of diffusivity in this material and the agreement found by M-10 and T-2 it appears likely that the suggested interstitialcy mechanism leading to a curved plot of  $\log D$  versus  $(1/T)$  is appropriate

and that the values for  $\text{Ag}^+$  diffusivity can be represented by:

$$D_{\text{Ag}^+}(\text{AgBr}) = 680 \exp (-1.10/kT) [345^\circ < T < 410^\circ\text{C}] \quad (6a)$$

$$D_{\text{Ag}^+}(\text{AgBr}) = 57 \exp (-0.97/kT) [250^\circ < T < 345^\circ\text{C}] \quad (6b)$$

$$D_{\text{Ag}^+}(\text{AgBr}) = 1.3 \exp (-0.79/kT) [140^\circ < T < 250^\circ\text{C}] \quad (6c)$$

Figure 4 presents the diffusion coefficient for bromine measured on AgBr single crystals. No more recent data are available.

#### Silver Iodide

The diffusivity of silver in the three common polymorphs of Ag I is presented in Figures 5 and 6. It may be noted that the observed data of Jordan and Pochon (J-24) are for pressed powder. The data of Zimen et al (Z-1) are for polycrystalline material and were performed with a short lived isotope of  $\text{Ag}^{111}$  which most probably makes the data somewhat less accurate. It is interesting to note that the activation energy for silver diffusion is lower in the iodide than in the other halides and that the cubic  $\gamma$ -Ag I has a smaller activation energy than the hexagonal  $\beta$ -Ag I. Moreover, the diffusivities are exceptionally high as compared to the other silver halides at the same temperature. Conductivity has been measured by Tubandt (T-12) and by Lieser (L-9) and is reported in the "Physics Handbook". This data was used to find diffusivity from the Einstein relation. The reported values of activation energy for Lieser's work is incorrectly presented in the "Handbook of Physics". Moreover, the data shows a strong temperature dependence of the activation energy. Therefore

Lieser's original data was used to prepare the curve shown in Figure 5. The strong curvature is rather unusual but has recently been substantiated on  $\beta$ Ag I single crystals (L107). These latter data also show a much lower conductivity than the earlier results (L-9). Apparently there is a strong grain boundary contribution in the polycrystalline material. The other significant feature of the conductivity data is that the calculated diffusivities are lower than the observed values. In view of the several different investigations this seems to be a real effect and suggests a possible ring mechanism for diffusion of Ag in  $\beta$  Ag I. The curvature observed by Lieser (L-9) and by Lakatos and Lieser (L107) has been explained (L107) in terms of lattice distortion and movement of interstitials through the saddle point. It must, however, be considered that the curvature and lower values of  $D_{calc}$  than  $D_{obs}$  are both consistent with impurity effects. There is a need for much more work in this area before a definitive understanding of diffusion in this material is forthcoming. Tentatively the observed values reported by Ziman et al (Z-1) may be used but single crystal diffusivity data for Ag are needed. Crystallographic work on the  $\beta \rightarrow \gamma$  transition and the possibility of derivative structures and superlattice ordering of defects in these phases is also needed so that the theory of the diffusivity and conductivity may be clarified.

The self diffusion coefficient for iodine in  $\beta$ -Ag I single crystals was also measured by Lakatos and Lieser (L107). The diffusivity along the c axis shows unusual behavior decreasing from about  $10^{-13}$  to about  $3 \times 10^{-14}$  between 50°C and 140°C. The diffusivity along the a axis rises with temperature in the normal fashion but has a very low activation energy of only about 0.3ev.

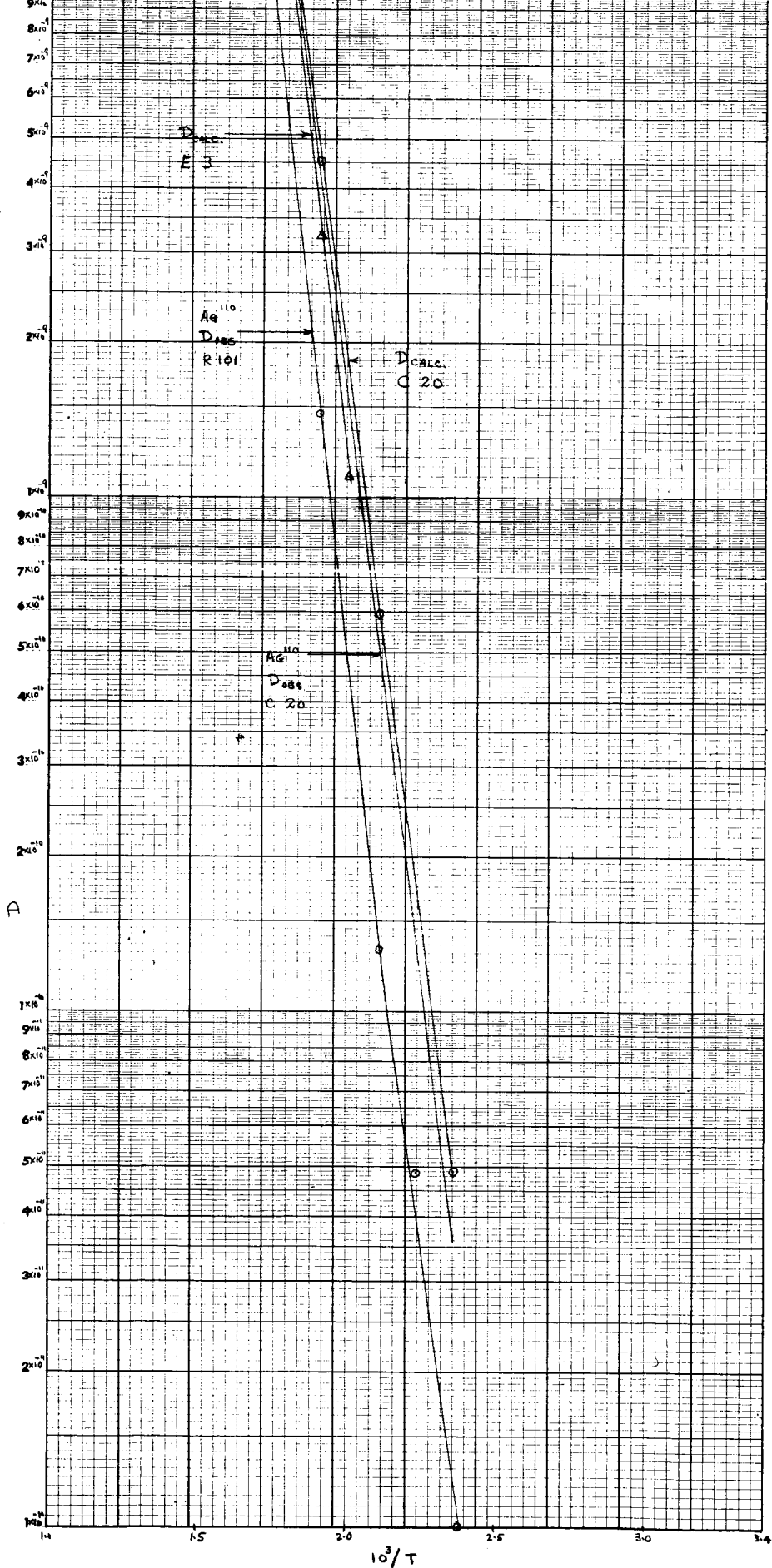
Figure 6 plots the diffusivity of silver in the high temperature  $\alpha$  phase. The only measured value is for a compact of pressed powders (J-24) and over a rather small range of temperature. The calculated values were obtained using the thermal expansion data of Lieser (L101). The excellent agreement of the Tubandt (T-12) and Lieser (L-9) data suggest that these conductivity data are probably reliable. In view of the fact that the diffusion coefficient was measured on a pressed powder compact and that the structure of Ag I (with 42 interstitial sites per unit cell (H101) probably indicates a simple interstitial mechanism, it is recommended that the diffusivity obtained from the Einstein relation be tentatively accepted:

$$D_{Ag} + (\alpha \text{ Ag I}) = 3.55 \cdot 10^{-4} \exp \left( \frac{-0.105}{kT} \right) \quad (7)$$

Figure 7 shows the diffusivity of I in Ag I. The excellent agreement of data obtained by Nölting (N101) with earlier results (J-28) suggest adoption of these values. The pressed powder results of Jordon and Pochon (J-24) are somewhat low and should probably be discounted. These low values obtained by them for anion diffusivity are particularly significant in view of the decision above to similarly discount their results for cation diffusivity in favor of diffusivity obtained from conductivity data.

K&E SEMI-LOGARITHMIC 46 4850  
2 CYCLES X 60 DIVISIONS  
MADE IN U.S.A.  
KEUFFEL & ESSER CO.

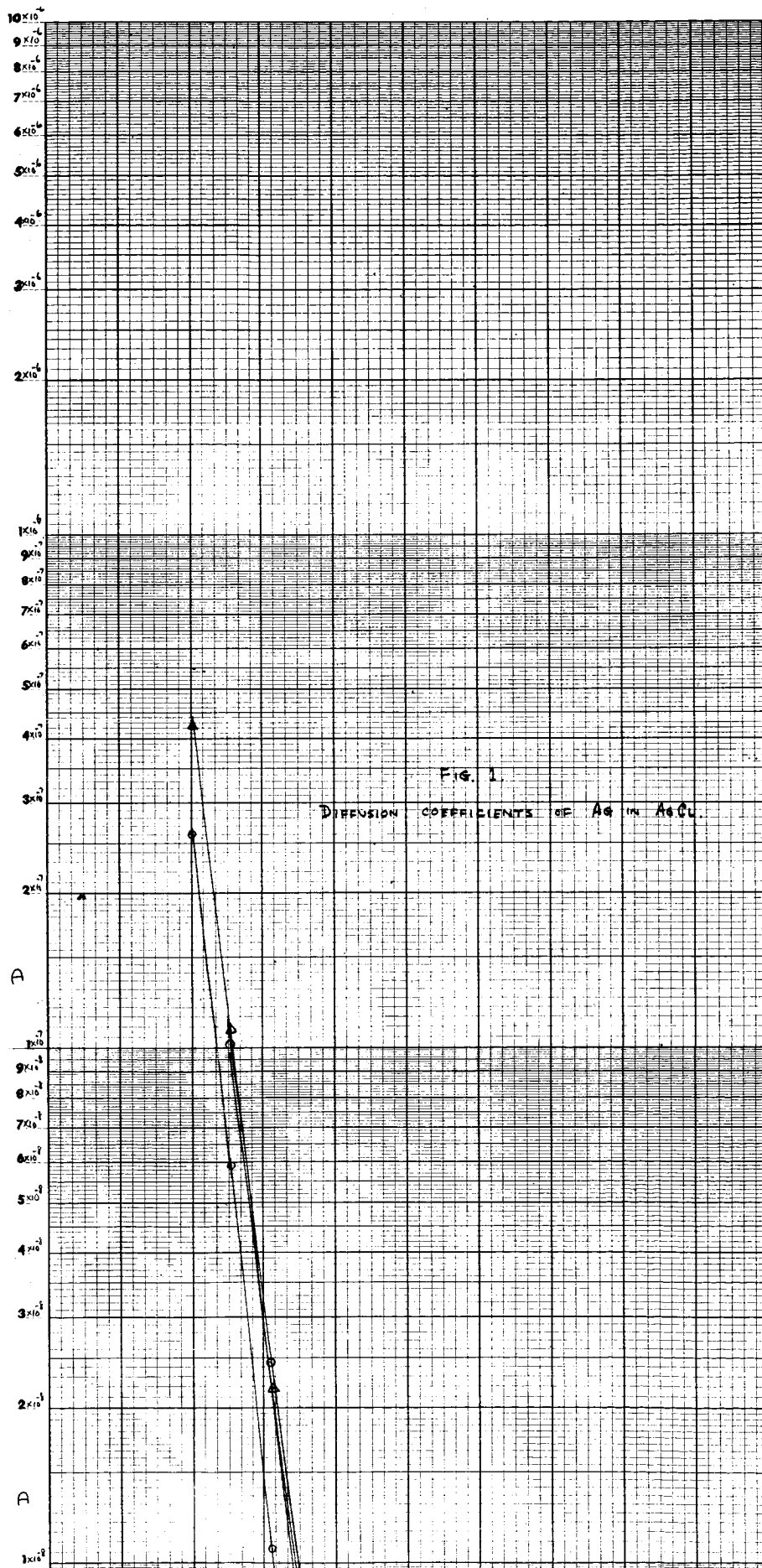
K&E SEMI-LOGARITHMIC 46 4850  
2 CYCLES X 60 DIVISIONS  
MADE IN U.S.A.  
KEUFFEL & ESSER CO.

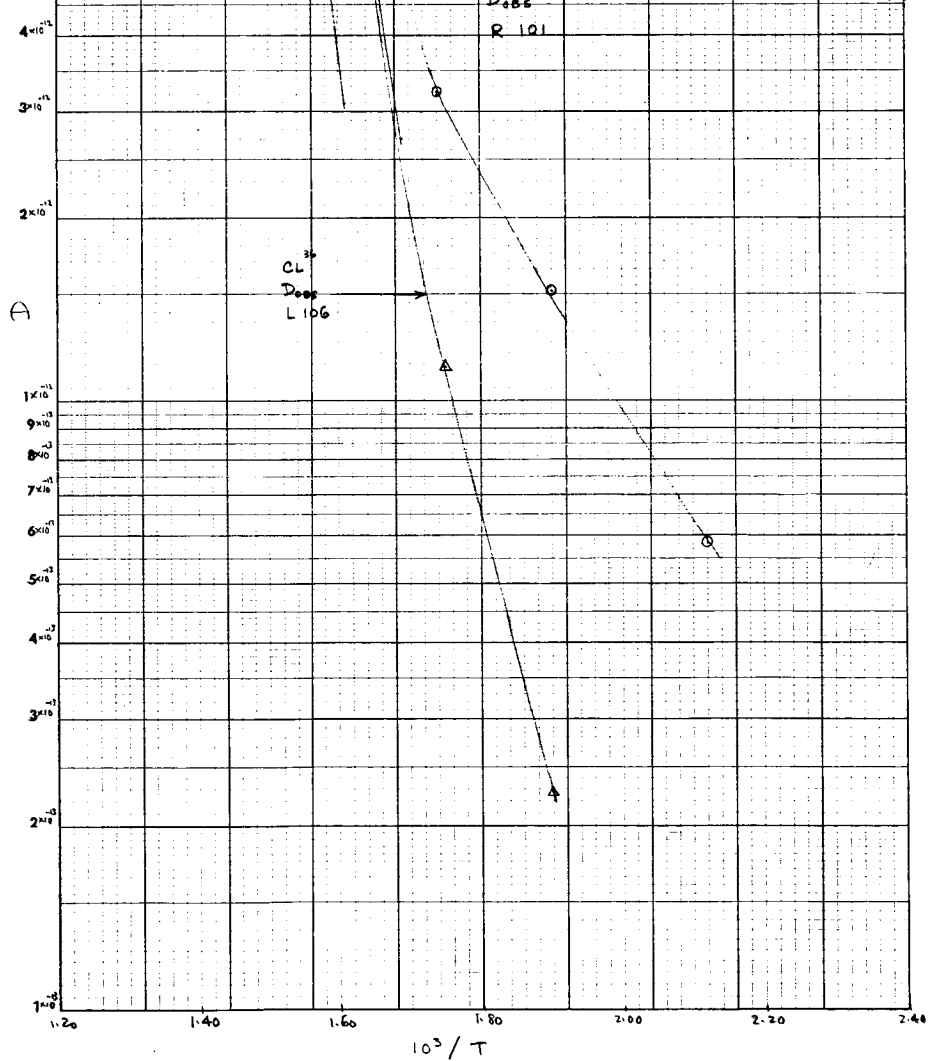


1

2

46 4850  
SEMI-LOGARITHMIC  
2 CYCLES DIVISIONS  
KEUFFEL & ESSER CO.

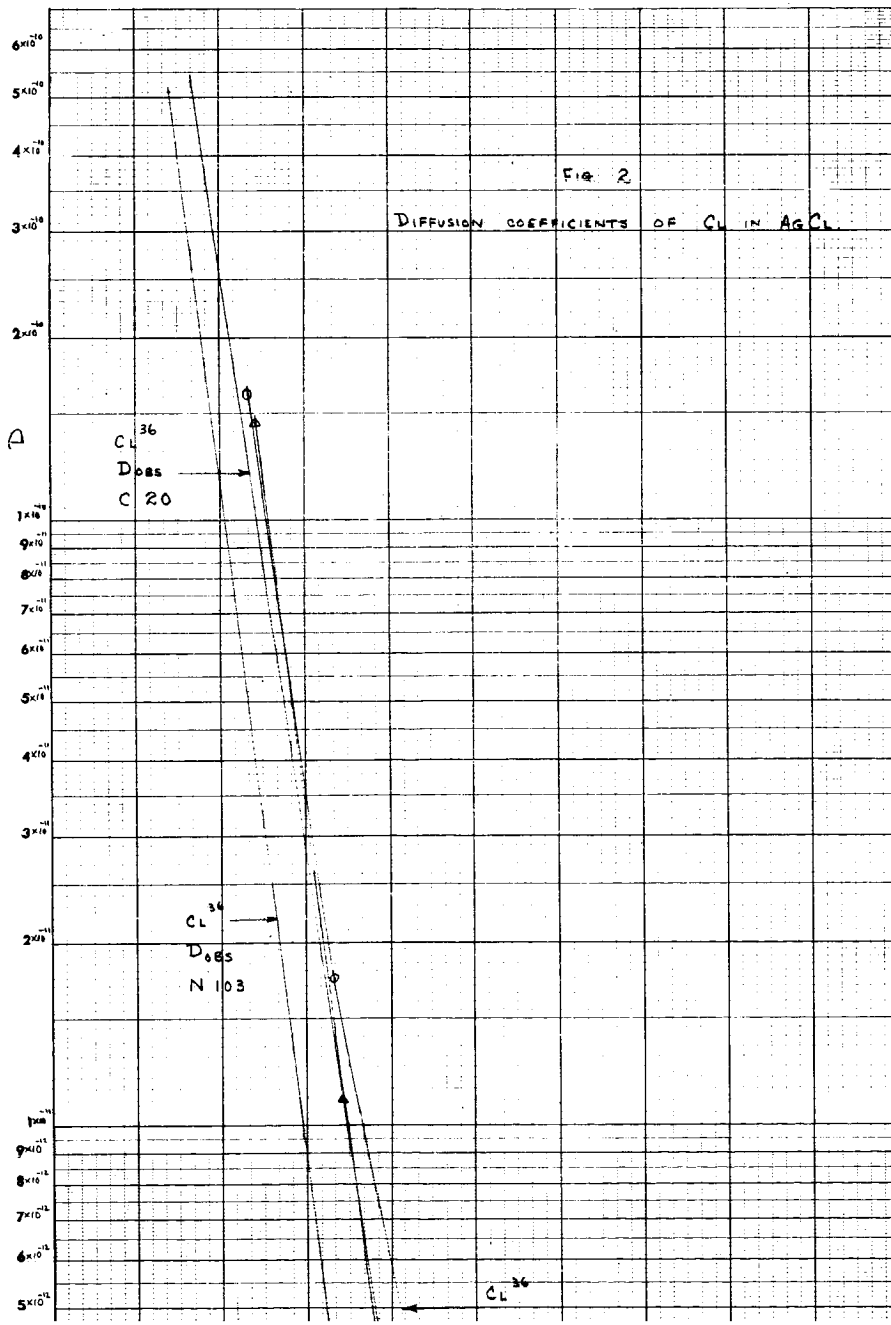





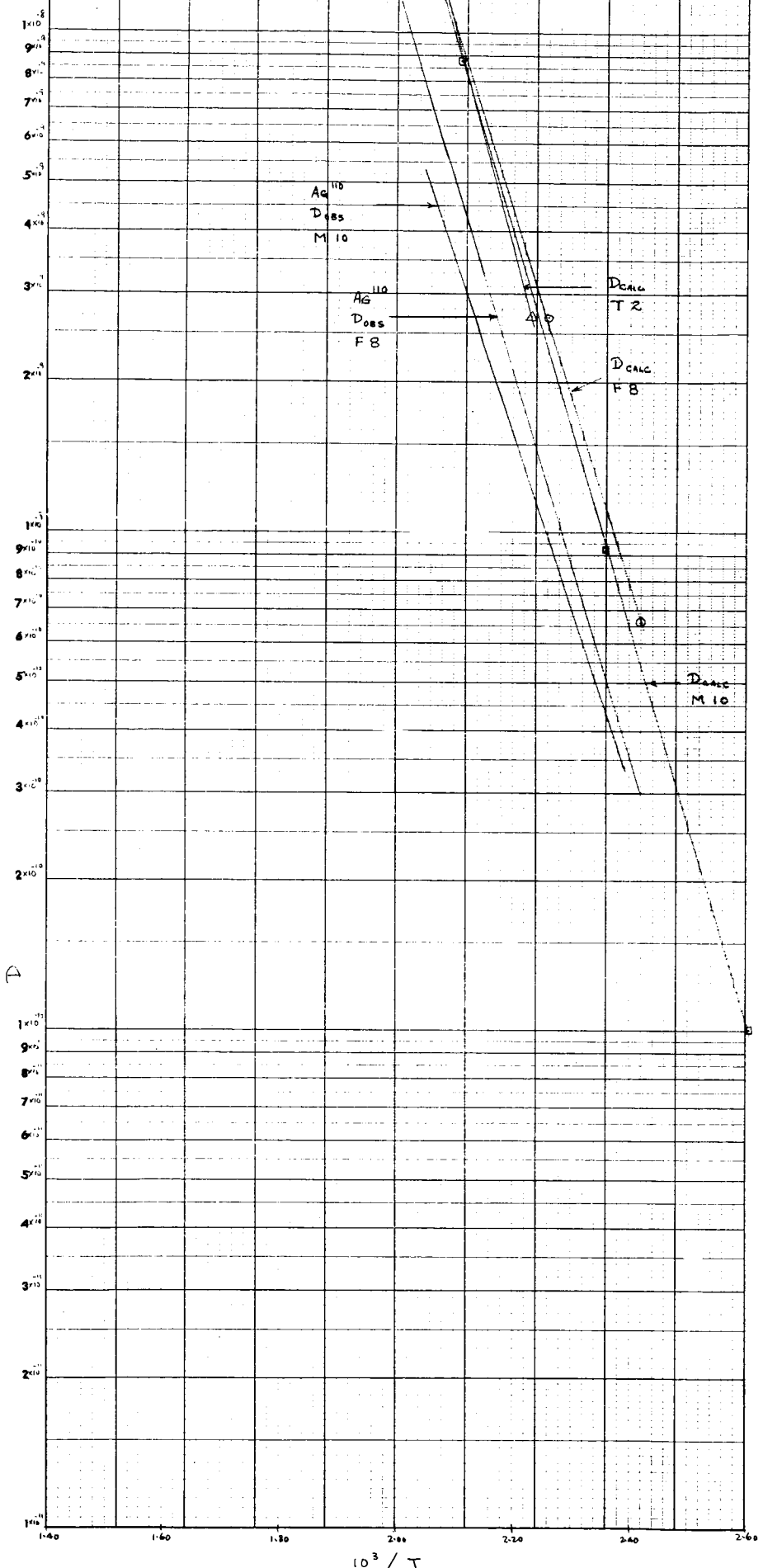


2

KE SEMI-LOGARITHMIC 46 4850  
2 CYCLES X 60 DIVISIONS  
KEUFFEL & ESSER CO.



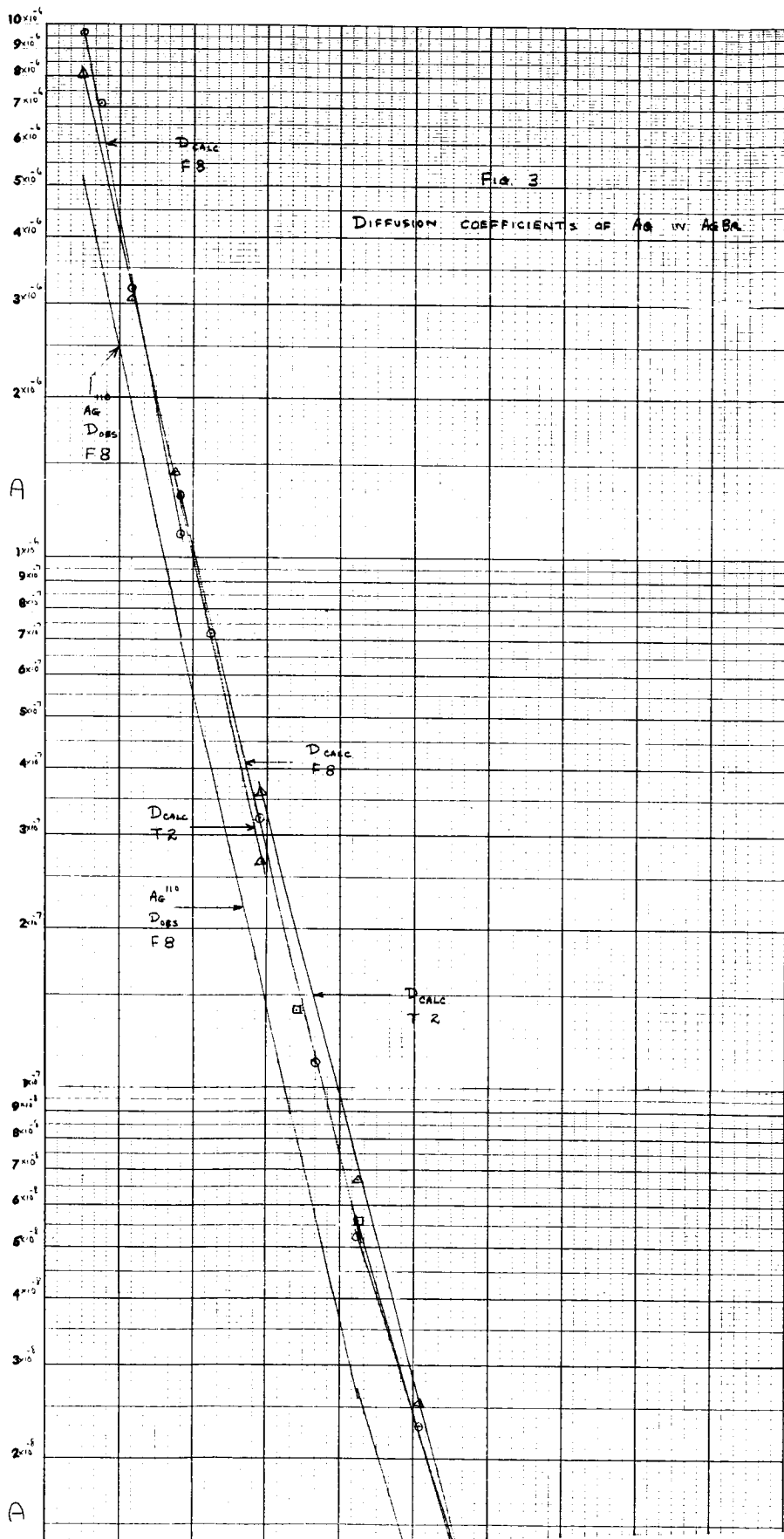

 SEMI-LOGARITHMIC 46 4850  
 KEUFFEL & ESSER CO.




 SEMI-LOGARITHMIC 46 4850  
 KEUFFEL & ESSER CO.

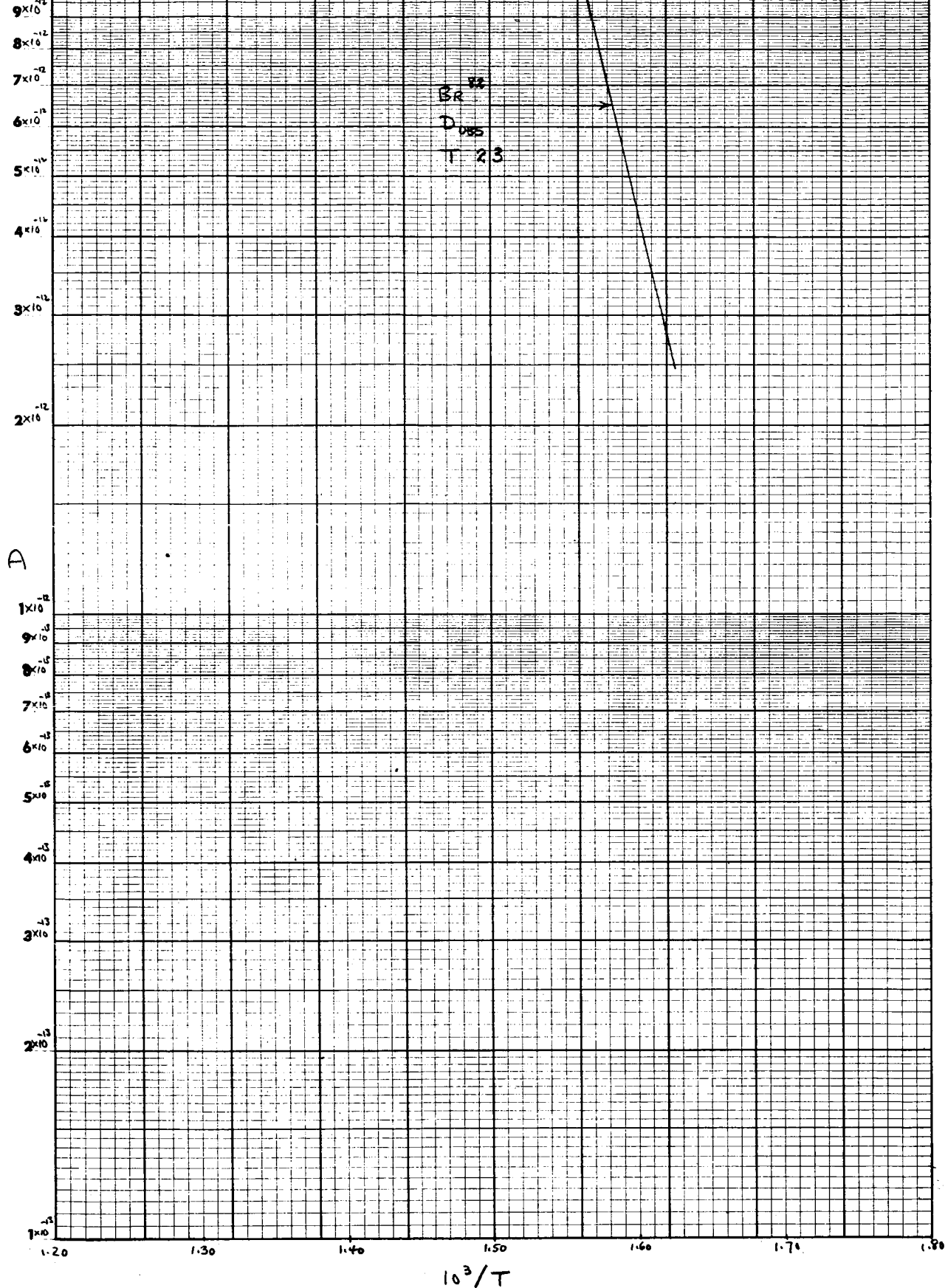
$10^3 / T$

SEMILOGARITHMIC 46-4850  
 DIFFUSION COEFFICIENTS  
 KEUFFEL & ESSER CO.

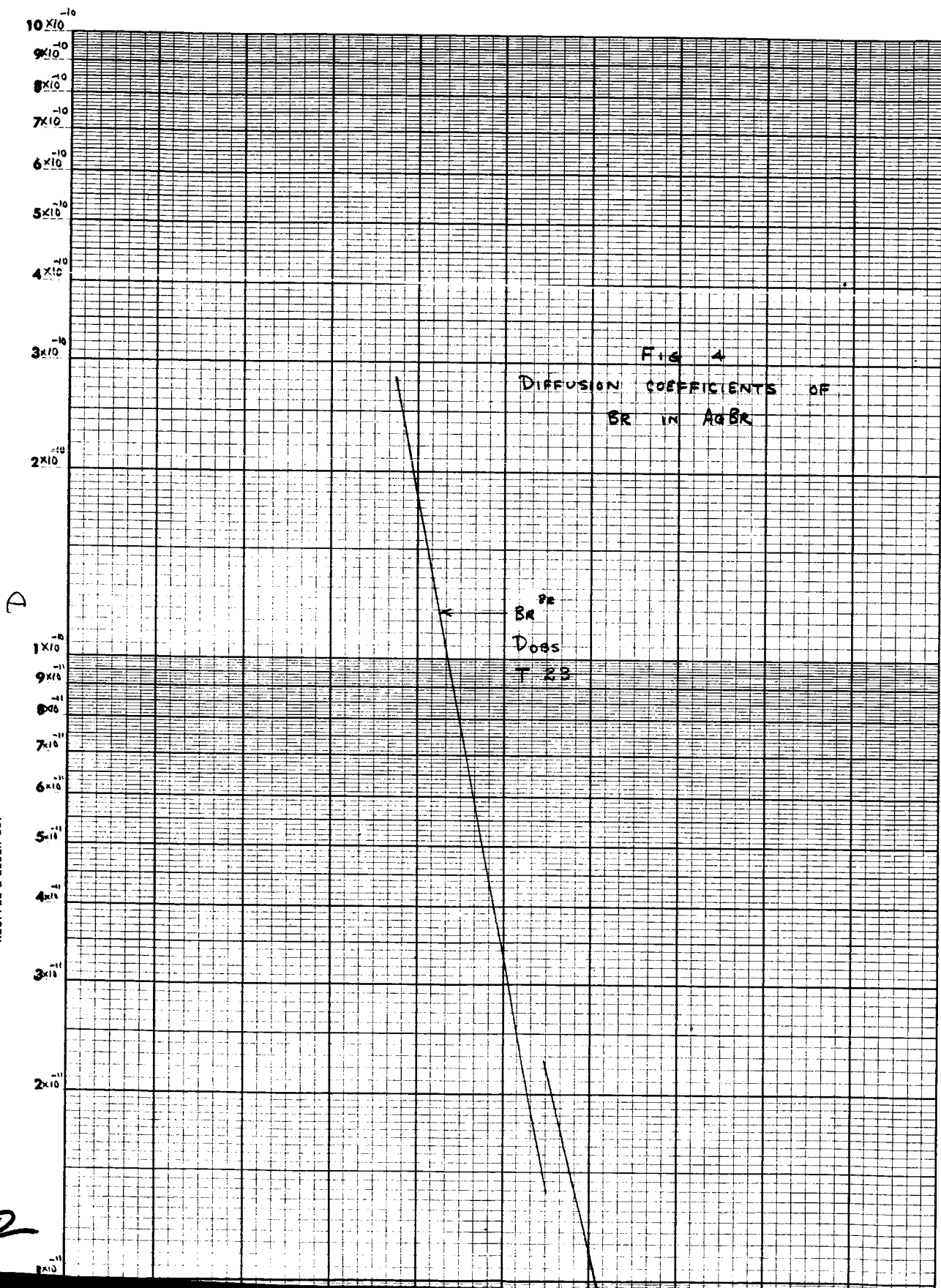


2

**K&E** SEMI-LOGARITHMIC 46 4850  
 2 CYCLES X 60 DIVISIONS  
 MADE IN U.S.A.  
 KEUFFEL & ESSER CO.

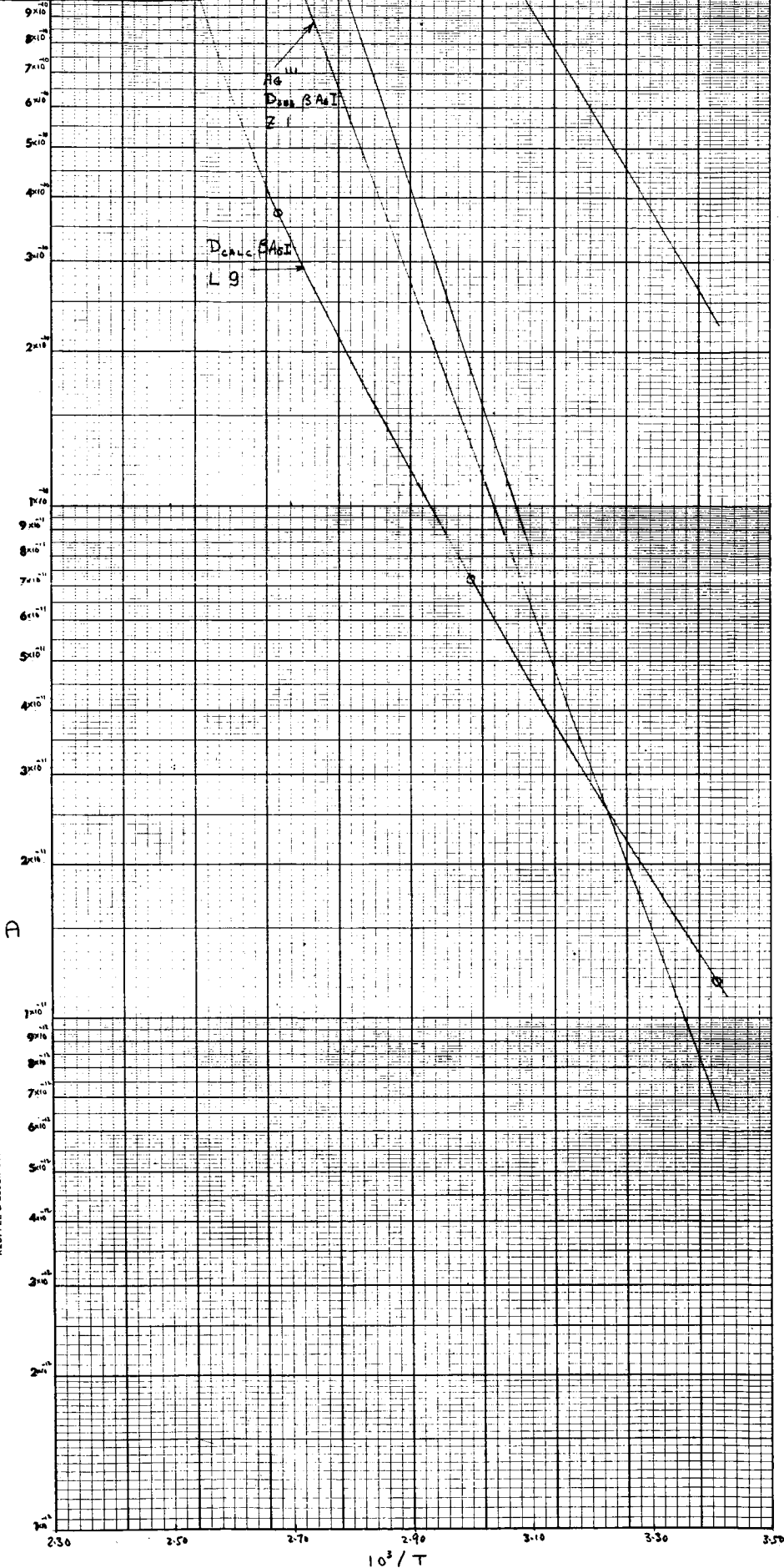


2



K-E SEMI-LOGARITHMIC 46 4850  
2 CYCLES X 60 DIVISIONS  
MADE IN U.S.A.  
KEUFFEL & ESSER CO.

K-E SEMI-LOGARITHMIC 46 4850  
2 CYCLES X 60 DIVISIONS  
MADE IN U.S.A.  
KEUFFEL & ESSER CO.



K&E SEMI-LOGARITHMIC 46 4850  
100 DIVISIONS  
 2 CYCLES X 60 DIVISIONS  
 KEUFFEL & ESSER CO.

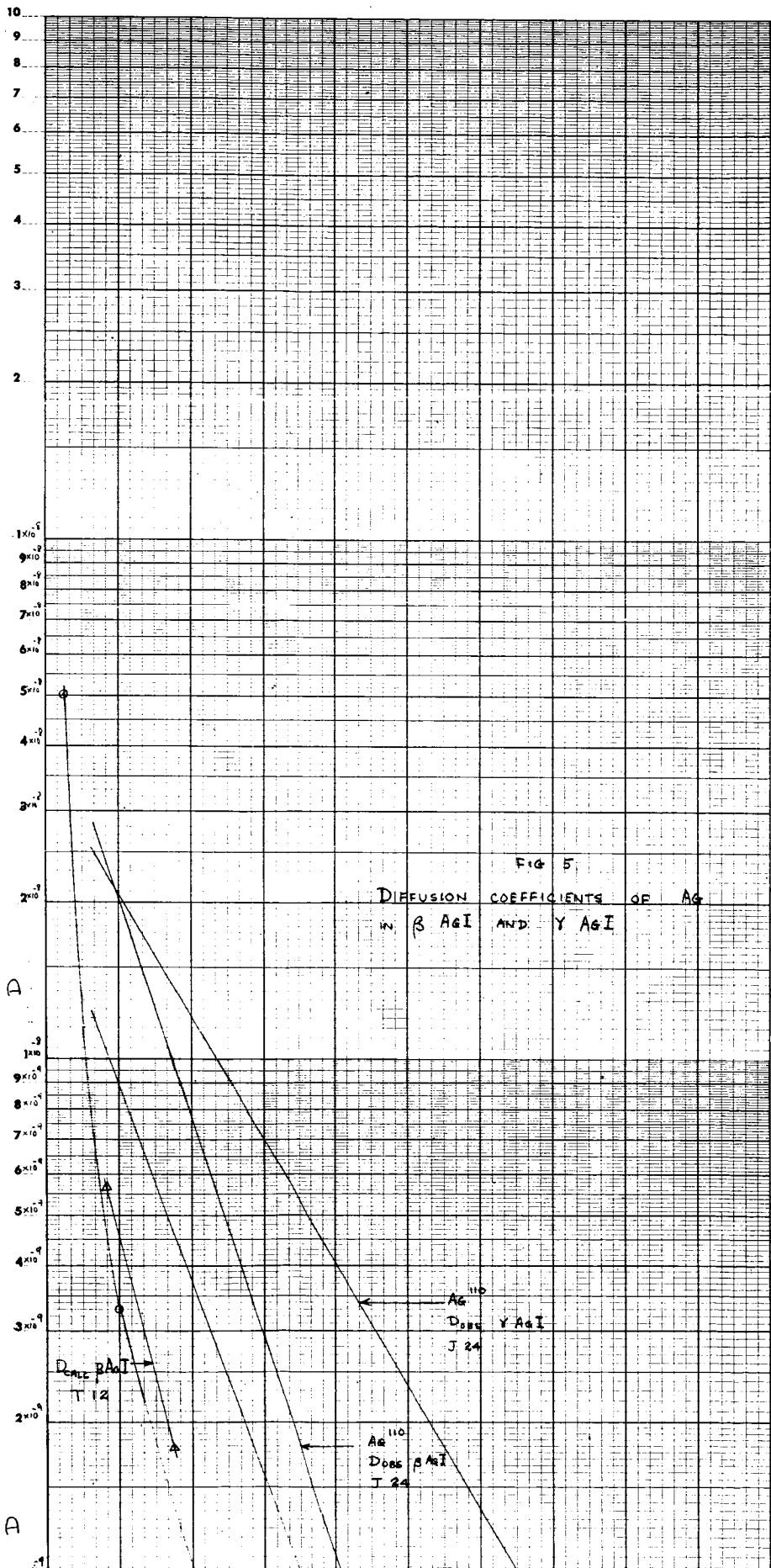


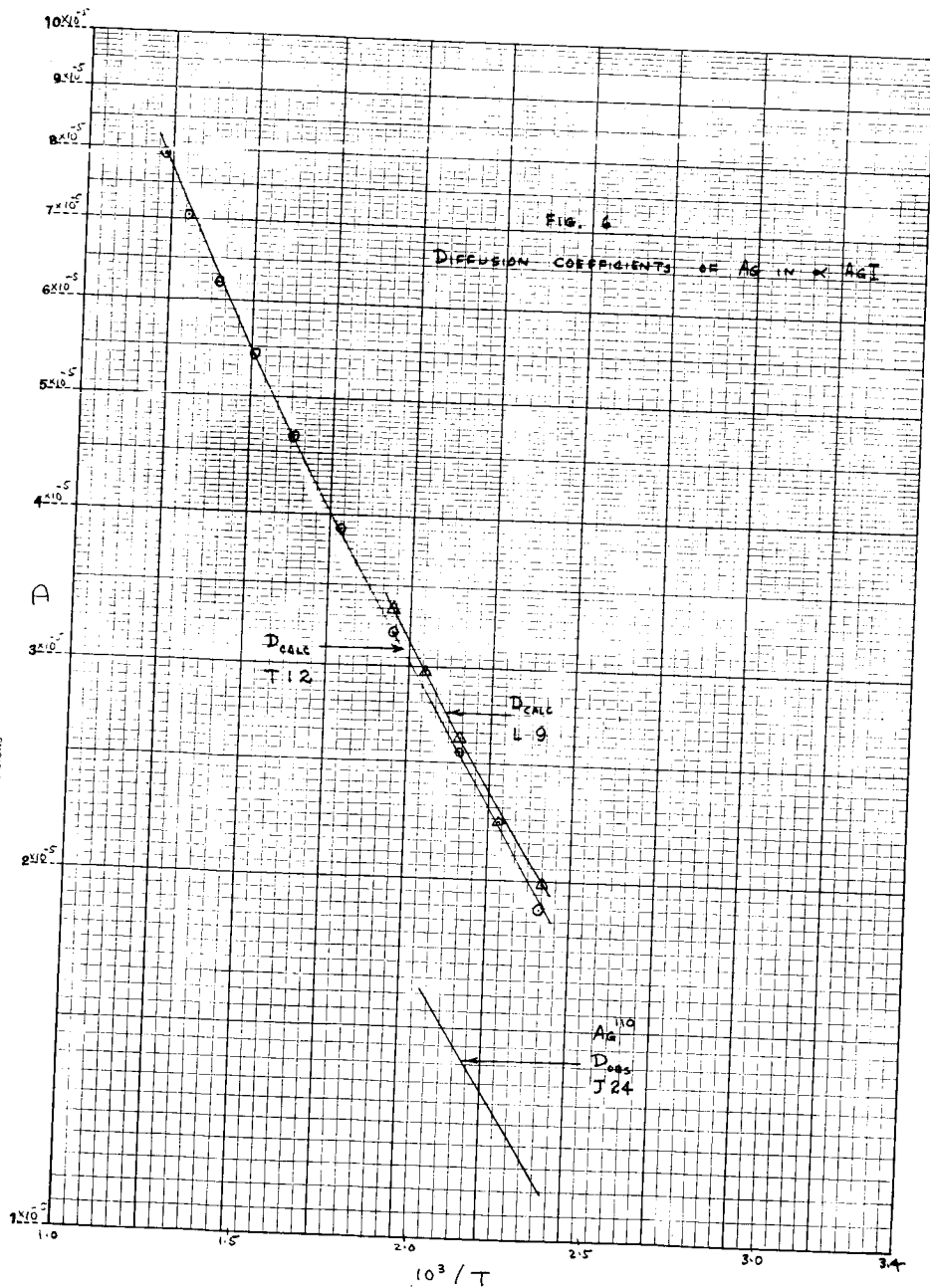
FIG 5  
 DIFFUSION COEFFICIENTS OF Ag  
 IN  $\beta$  AgI AND  $\gamma$  AgI

$D_{0.05} \beta \text{ AgI}$   
 $T 12$

$\text{Ag}_{110}$   
 $D_{0.05} \gamma \text{ AgI}$   
 $T 24$

$\text{Ag}_{110}$   
 $D_{0.05} \beta \text{ AgI}$   
 $T 24$

✓





**K&E** SEMI-LOGARITHMIC 46 4850  
 2 CYCLES X 60 DIVISIONS  
 MADE IN U.S.A.  
 KEUFFEL & ESSER CO.

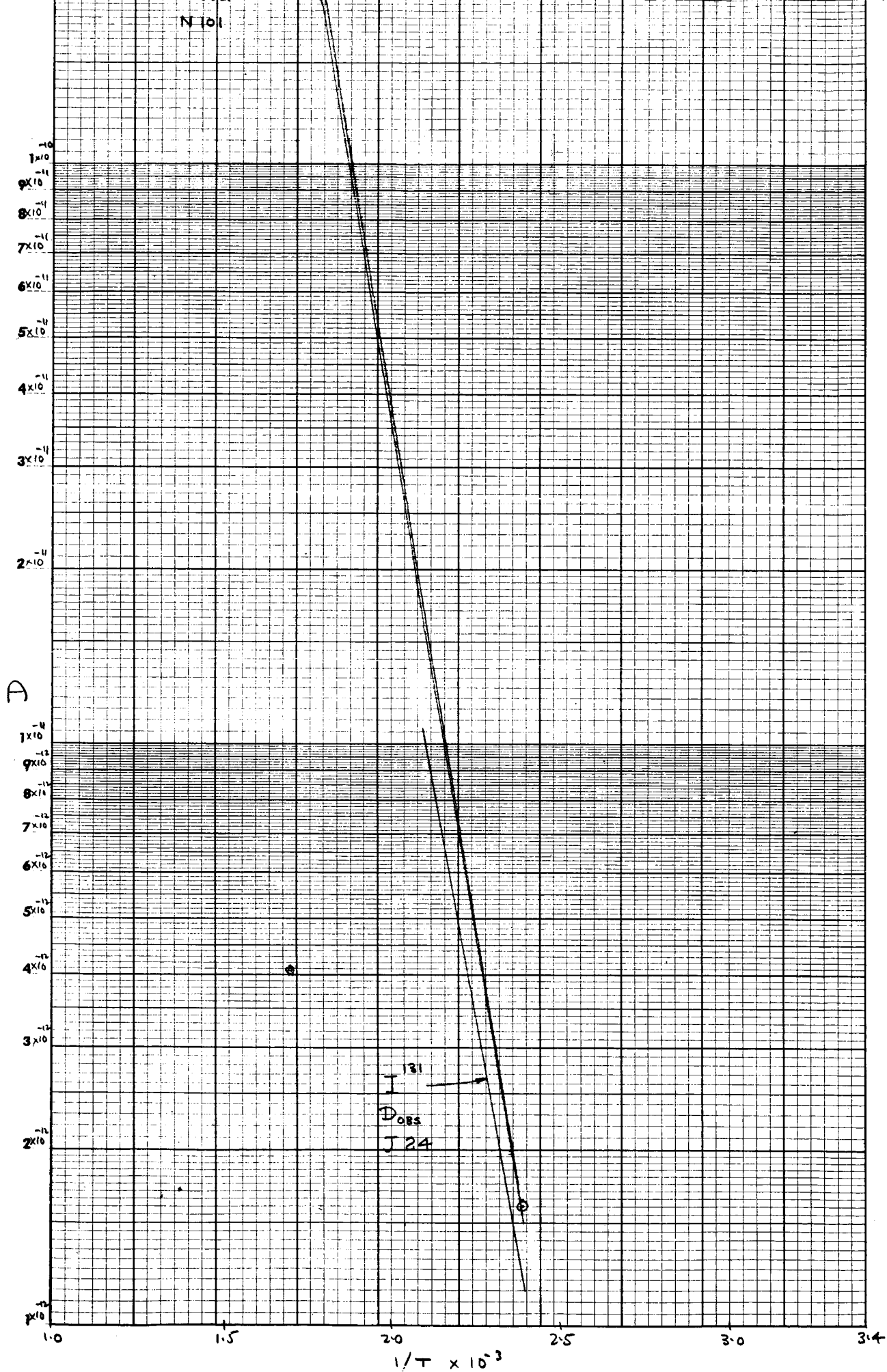
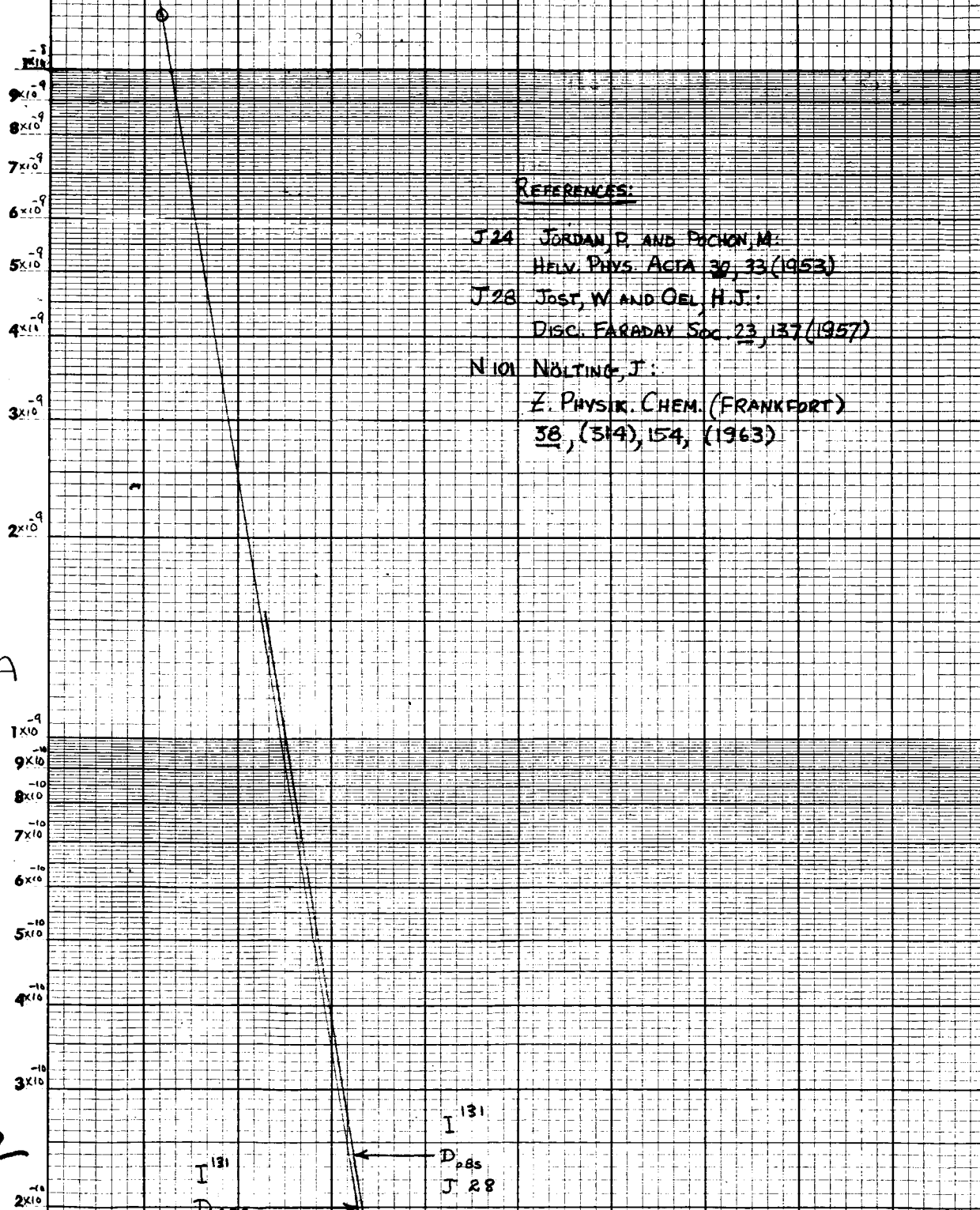


Fig. 7 DIFFUSION COEFFICIENTS OF I<sup>-</sup> IN  $\alpha$ -AgI



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## VII TURBULENT TRANSPORT DATA

### I. Introduction

The transport of momentum, heat and chemical species in turbulent flow fields plays an important role in many areas of science and engineering. Research on turbulent transport phenomena is being actively pursued at a number of laboratories. In nearly every case the work is directed toward a rather narrow and specific goal and deals with limited ranges in properties and geometry of the systems being studied. The objective of the present program is to compile and evaluate the results being obtained in as broad a manner as possible.

As a consequence of the statistical nature of turbulence and the role of geometry the survey has been sub-divided into confined and free flow fields. Some of the types of transport in confined flow are: heat, mass and momentum transport in conduits; porous media, and boundary layers. Free or unconfined flow includes: jets and streams issuing into large bodies; meteorological and oceanographic flows. Work in these areas is discussed in Section III.

Only systems for which the molecular transport coefficients are well established are being considered. Excluded from the survey at least for the present are: non-Newtonian fluids; systems at extremely high or low temperatures; magnetohydrodynamic systems; and, very low pressure gases.

In the analysis of practical problems dealing with a complex phenomena such as turbulence it is necessary at the onset to establish the degree of detail required. For example one can measure and correlate data on heat transfer from fairly simple experiments. This information will be of value in future predictive and design work, but at no time in the development of this fund of information was it necessary to measure the values of the Reynolds stress and temperature-velocity correlations. This approach has been and is being

carried out in nearly all practical problems dealing with turbulent flow fields and we now have quite reliable empirical equations for quantities such as friction, drag, heat and mass transfer coefficients covering a wide variety of geometries and flow conditions.

There are a number of problems, such as the turbulent flow of chemically reacting fluids, where a knowledge of the nature and structure of the flow field is required. In addition quantities such as the Reynolds stresses and correlation coefficients are needed the integration of the point equations of change to obtain integral quantities such as friction and heat transfer coefficients. Thus a better understanding of the nature of turbulence will provide a better basis for the correlation of the design type of data mentioned above.

A brief summary of the more frequently used statistical properties of turbulent transport is presented in Section II. Discussion of triple and other correlations is omitted from Section II. The double correlations arise because of the basic nonlinearity of the equations of change and they have been measured and reported by a number of investigators. The equations for the double correlations introduce triple correlations, and so on. In fact the system of correlation equations does not close<sup>1</sup>. Since there have been few measurements of triple and other correlations they have been excluded at least for the present<sup>2</sup>.

NASA provides two abstraction, and indexing services that are of considerable value in this work. The International Aerospace Abstracts (IAA) covers published literature and the Scientific and Technical Aerospace Reports (STAR) covers

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<sup>1</sup> A. J. Favre, A. S. M. E., J. Applied Mech., 32, 241 (1965)

<sup>2</sup> Deardorff, J. W., Proc. Heat Trans. Fluid Mech. Inst., 16, Stanford Univ. Press (1965)

reports and other unpublished information. An indication of the activity in the area of turbulent transport may be obtained from the number of abstracts listed in these services during recent years.

YEAR	STAR	IAA
1963	93	232
1964	210	276
1965	219	356

As shown above there were 356 papers and articles dealing with turbulence, trubulent effect, turbulent air current, turbulent boundary layer, turbulent diffusion, turbulent flow, turbulent heat transfer, turbulent jet, turbulent mixing and turbulent wake abstracted in IAA during 1965.

We estimate that about 80% of the new literature on turbulence is being covered by these NASA abstracts. It has also been our experience that over half the data contained in the reports covered by STAR appears again as published information a year or two latter in IAA. Finally, it seems that slightly less than 10% of these papers and reports contain new experimental data on turbulent transport.

Some measure of the magnitude of the literature prior to 1960 may be obtained by considering the number of references cited in two texts that are oriented more toward the experimental side of turbulent flow. Hinze<sup>3</sup> cites over 300 references of which about one fourth contain experimental results. In flow Schlichting<sup>4</sup> cites a little over 440 references.

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<sup>3</sup> J. O. Hinze, Turbulence, McGraw-Hill, (1959)

<sup>4</sup> H. Schlichting, Boundary Layer Theory, McGraw-Hill 2nd (1960)

## II. TRANSPORT CHARACTERISTICS OF TURBULENT FLOW FIELDS

This section summarizes the nomenclature and properties of turbulent transport with regard to the quantities which must be measured to adequately describe each system. The need for such a section arises from the preliminary finding that in a number of instances not all the characteristic quantities necessary for the determination of turbulent transport properties are reported by the investigators. This is particularly true in unconfined flow (jets and streams, meteorological and oceanographic flows) where the primary interest is in structure of the flow rather than transport phenomena.

Turbulent transport occurs in flow fields which are characterized by random fluctuations of the velocity, concentration and temperature. At low velocities streamlines are usually regular and well defined. However, at higher velocities instabilities develop resulting in fluctuations characteristics of turbulence. The particle of fluid exhibiting such motion, the eddy, resembles the spiral galaxies in photographs of the heavens. The majority of the eddies involved in the heterogeneous motion known as turbulence are small and exhibit their presence through bulk action in the phenomena of diffusion and energy dissipation.

Only systems for which the molecular transport coefficients are well established are being considered. Excluded from the survey are: non-Newtonian fluids; systems at extremely high or low temperature; magnetohydrodynamic systems; very low pressure gases; and, systems composed of more than two chemical species.

The equations that describe the transfer rates and profiles

are known as the equation of change.<sup>1,2</sup> These equations are general conservative statements for the conservation of momentum, energy and chemical species. Turbulent flows form a special class of solutions to the equations in which the dependent variables (velocity, temperature, etc.) are not simple functions of space and time. Consequently, recourse is usually made to describing the variables by probability laws.

#### A. Equations of Change

In applying the equations of change to turbulent flow the large majority of investigators employ the procedure, introduced by Reynolds, of resolving the dependent variable into the sum of a time average value and a fluctuating value. After this substitution the equations are time smoothed. The resulting equations resemble the original equations expressed in terms of the time average except that they contain certain time averaged products of fluctuating quantities. These terms represent the turbulent contribution.

##### 1. Time Smoothing

The time smoothing is carried out by applying the mean value theorem

$$\bar{f} = \frac{1}{T} \int_{t_0}^{t_0+T} f \, dt \quad (1)$$

where  $\bar{f}$  is time smoothed by taking the average over a time interval,  $T$ , large with respect to the time of turbulent oscillations but small with respect to possible long term transients.<sup>1</sup>

<sup>1</sup>Bird, R. B., W. E. Stuart and E. N. Lightfoot, Transport Phenomena, pp. 153, 375 and 626, Wiley (1960).

<sup>2</sup>Hinze, J. O., Turbulence, p. 13, McGraw-Hill (1959).

In keeping with the general procedure of resolving  $f$  into an average and a fluctuating component equation (1) may be written

$$\bar{f} = \frac{1}{T} \int_0^{t+T} (f + f') dt = \bar{f} + \bar{f'} \quad (2)$$

and it follows that

$$\bar{\bar{f}} = \bar{f} \quad (3a)$$

$$\overline{f'} = 0 \quad (3b)$$

Because the product of functions frequently occurs in the equation of change it is convenient to list several additional rules<sup>3</sup> of averaging which are simply properties of the integration.

$$\overline{\frac{\partial f}{\partial s}} = \frac{\partial \bar{f}}{\partial s} \quad (3c)$$

$$\overline{f \cdot g} = \bar{f} \cdot \bar{g} \quad (3d)$$

$$\overline{f \cdot g} = \bar{f} \bar{g} + \overline{f' g'} \quad (3e)$$

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<sup>3</sup>Schlichting, H., Boundary Layer Theory, p. 460, McGraw-Hill (1960).

## 2. Equation of Motion

Application of Newton's second law of motion to flow of fluids leads to Cauchy's equation of motion<sup>4</sup> when the fluid is treated as a continuous medium,

$$\frac{\partial}{\partial t} (\bar{N}) = - [\nabla \cdot (\bar{N} \bar{N})] - \nabla p - [\nabla \cdot \bar{T}] + \rho \bar{g} \quad (4)$$

considering only gravitational field forces. The first term on the right represents the rate of convected momentum and the third the rate of viscous momentum transfer.

Consideration of the stress tensor<sup>1,4</sup> for Newtonian fluids shows that for the conventional time smoothing no turbulent terms appear. The only turbulent contribution comes from time smoothing the dyadic product of  $\bar{N}$  and  $\bar{N}$  in the convected momentum rate of equation (4). This results in the Reynolds stresses representing the turbulent contribution to the momentum flux. They are:

$$\bar{T}^{(c)} = \overline{(\bar{N} \bar{N})} \quad (5a)$$

for compressible flow and

$$\bar{T}^{(c)} = \rho \overline{\bar{N} \bar{N}} \quad (5b)$$

for incompressible flow.

When the viscosity and density are constant the equation of motion becomes the Navier-Stokes equation. The time

<sup>4</sup>Aris, R., Vectors, Tensors, and the Basic Equation of Fluid Mechanics, p. 102, Prentice-Hall (1962).



smoothed form of the Navier-Stokes equation is:

$$\rho \frac{\partial \bar{\mathbf{u}}}{\partial t} = - (\nabla \cdot \bar{\mathbf{N}} \bar{\mathbf{u}} - \nabla \bar{P} - \mu \nabla^2 \bar{\mathbf{u}} - \rho \nabla \cdot \bar{\mathbf{u}} \bar{\mathbf{u}} + \rho \bar{\mathbf{u}} \nabla \cdot \bar{\mathbf{u}}) \quad (6)$$

Equation (6) will be the starting point for most fluid mechanic problems. The main exception being gases in sonic and supersonic flow.

### 3. Energy Equation

The first law of Thermodynamics for an open, unsteady state system is the basis for the energy equation. When thermal energy only is considered for a fluid of constant molecular properties the time smooth energy equation becomes:

$$\rho C_p \frac{\partial \bar{T}}{\partial t} = - \rho C_p \nabla \cdot \bar{\mathbf{u}} \bar{T} - \rho C_p \nabla \cdot \bar{\mathbf{u}}' \bar{T} + k \nabla^2 \bar{T} + \bar{q} \quad (7)$$

The viscous dissipation term has been omitted from equation (7) since it only appears in certain lubrication and high velocity gas flow cases.<sup>1,5</sup> However, the viscous dissipation term does make a turbulent contribution in those instances.

### 4. Continuity Equations

The principle of conservation of mass leads to the continuity equation. The time smoothed form for an incompressible fluid is

$$\nabla \cdot \bar{\mathbf{u}} = 0 \quad (8a)$$

<sup>5</sup>Goldstein, S. (ed.), Modern Developments in Fluid Dynamics, 2 p. 611, Oxford University Press, London (1938)

and the fluctuating component obeys the expression:

$$\nabla \cdot \vec{N}' = 0 \quad (8b)$$

Conservation of chemical species leads to the time smoothed diffusion equation. For an incompressible fluid of constant diffusivity and a first order chemical reaction:

$$\begin{aligned} \frac{\partial \bar{C}_A}{\partial t} = & - \nabla \cdot \bar{N} \bar{C}_A - \nabla \cdot \overline{N' C'_A} \\ & + D_{AB} \nabla^2 \bar{C}_A + k_A \bar{C}_A \end{aligned} \quad (9)$$

If the reaction rate is second order or higher there are turbulent contributions from the rate expression. Equation (9) contains only the ordinary molecular diffusion flux arising from Fick's law. Other types of molecular diffusion such as thermal, pressure and forced (ionic) must be included when necessary.

## B. Properties of Turbulence

The time smoothed products of fluctuating quantities contained in equations (6, 7 and 9) represent the turbulent contribution to the transport. These expressions have been interpreted in terms of statistical and semi-empirical models. Only those properties of turbulence that directly relate to the transport are considered.

### 1. Correlation Coefficient

The statistical treatment of the fluctuating quantities leads to the introduction of a correlation between these quantities. The correlation coefficients, based on the Eulerian view, involve the product of the fluctuating

quantities at two different points in space separated by a distance  $r$ .

$$R_{fg}(r) = \frac{f(r)' g(r)'}{\sqrt{f(r)'^2} \sqrt{g(r)'^2}} \quad (10)$$

The intensity of turbulence is a measure of the magnitude of the fluctuations about the mean value and is expressed as the root-mean-square value,  $\sqrt{f(r)'^2 / \overline{f(r)'}}$ . The magnitude of the fluctuations alone is not adequate in describing the turbulence. It is also necessary to specify a characteristic length which is a measure of the magnitude of turbulent eddies. This length is known as the scale of turbulence defined by:

$$L = \int_0^\infty R_{ff}(r) dr \quad (11)$$

At a point the correlation between components of the fluctuating velocity is given by:

$$R_{fg}(0) = \frac{f(0)' g(0)'}{\sqrt{f(0)'^2} \sqrt{g(0)'^2}} \quad (12)$$

The turbulent quantities in equation (6, 7, and 9) may be expressed in terms of the point correlation coefficient,

$$\overline{u_i' u_j'} = \sqrt{\overline{u_i'^2}} \sqrt{\overline{u_j'^2}} R_{ij} \quad (13a)$$

$$\overline{N_i' T'} = \overline{N_i'^2} \overline{T'^2} R_{iT} \quad (13b)$$

$$\overline{N_i' C'} = \overline{N_i'^2} \overline{C'^2} R_{iC} \quad (13c)$$

There is a distribution of eddy size and density in the flow field. Consequently, as in the molecular case, there are instances when one must treat the eddies individually and other instances where a large number may be treated as a continuum. The smaller eddies are more important in transport phenomena and serve to justify the use of time average point functions.<sup>6</sup>

## 2. Turbulent Transport Coefficients

An analogy has been established between turbulence and molecular theory. In this regard turbulent transport coefficients have been defined and used.

The turbulent transport coefficients may be defined in a manner similar to Newton's law of viscosity, Fourier's law of conduction and Fick's law of diffusion. The turbulent quantity in equation (6) may be expressed in terms of the time smoothed velocity and the eddy diffusivity of momentum,

$$\overline{N_i' N_j'} = - \epsilon_{ij} \left[ \nabla \overline{N_i} + (\nabla \overline{N_j})^+ \right] \quad (14a)$$

<sup>6</sup>Hinze, J. O., op. cit., p. 188.

Similarly the turbulent quantity in equation (7) may be expressed in terms of the time smoothed temperature and the eddy diffusivity of heat,

$$\overline{N'T'} = -\epsilon_h \cdot \nabla \bar{T} \quad (14b)$$

and the turbulent quantity in equation (9) in terms of the time smoothed concentration and the eddy diffusivity of mass is:

$$\overline{N'C'_x} = -\epsilon_D \cdot \nabla \bar{C} \quad (14c)$$

There exists some doubt as to the tensor nature of the turbulent transport coefficients.<sup>7</sup> From the rules of tensor multiplication the coefficients, as defined in equation (14), must be of even order (zero, second, etc.)

In simple flow, where the time average velocity is only in one direction, equation (14) takes on the Boussinesq form for the definition of the eddy coefficients.<sup>8</sup> If the mean velocity is  $\bar{N}_1$ ; equation (14) becomes:

$$\overline{N'_1 N'_2} = -\epsilon_L \frac{\partial \bar{N}_1}{\partial x_2} \quad (15a)$$

$$\overline{N'_2 T'} = -\epsilon_h \frac{\partial \bar{T}}{\partial x_2} \quad (15b)$$

$$\overline{N'_2 C'_x} = -\epsilon_D \frac{\partial \bar{C}}{\partial x_2} \quad (15c)$$

<sup>7</sup>Hinze, J. O., op. cit., pp. 25, 382

<sup>8</sup>Bird, R. B., op. cit, pp. 160, 379, 629

Consideration of equation (13, 14 and 15) indicate that the turbulent transport coefficients may be expressed in terms of the correlation coefficients, root-mean-square value of the fluctuating components and the mean gradients.

### 3. Phenomenological Approaches

Early investigators developed mechanistic concepts of the turbulent transport process. These are based mainly upon two hypotheses: (1) an analogy exists between the transfer of momentum or vorticity and the kinetic theory of gases; and, (2) that mixing lengths depend on the flow pattern. Some of the ideas of these workers merit consideration.

Prandtl developed an expression for momentum transfer in a fluid in terms of the mixing length:

$$\overline{(\rho \bar{u}'_1 \bar{u}'_2)} = - \rho l^2 \left| \frac{\partial \bar{u}_1}{\partial x_2} \right| \frac{\partial \bar{u}_1}{\partial x_2} \quad (16)$$

A similar result was obtained by Taylor from vorticity transport considerations. The mixing length is a function of position and is roughly proportional to the distance from a solid boundary.

Assuming that the flow patterns show geometrical similarity throughout the flow field, von Karman proposed that the Reynolds stresses have the form,

$$\overline{(\rho \bar{u}'_1 \bar{u}'_2)} = - \rho K^2 \left| \frac{\left( \frac{\partial \bar{u}_1}{\partial x_2} \right)^2}{\left( \frac{\partial^2 \bar{u}_1}{\partial x_1^2} \right)^2} \right| \frac{\partial \bar{u}_1}{\partial x_2} \quad (17)$$

where  $K$  is a universal constant whose value is about 0.4 for tube flow.

From dimensional considerations Deissler developed an expression for the flow behavior near a solid surface where Prandtl and von Karman's equations are inadequate,

$$\overline{v' u'} = - \left( \overline{v' u'} X_2 \right) \left\{ 1 - e^{-N X_2} \right\} \frac{d \overline{u}}{d X_2} \quad (18)$$

where  $X_2$  is the distance from the surface and  $N$  is a constant whose value is 0.124 for the tube flow.

Consideration of the wake flow behind objects lead Townsend<sup>9</sup> to suggest that the turbulent transport may be brought about by a gradient type diffusion resulting from the small-scale turbulence plus a convective contribution from large scale motions of eddies. He concludes that equation (15a) applies for momentum transfer. However, in place of (15b and 15c) he proposes,

$$\overline{v' u'} = - \epsilon \frac{d \overline{u}}{d X_2} + \overline{V' u'} \quad (19)$$

where  $V_2$  is the velocity of large scale eddy motion of magnitude comparable to in size to the width of the wake.

<sup>9</sup>Hinze, J. O., op. cit., p. 288.

### III Areas of Turbulent Transport

#### A. Introduction

Most flows which occur in practical applications are turbulent in that an irregular fluctuation or mixing motion is superimposed on the mainstream. The fluctuations are so hopelessly complex in detail that they seem to be inaccessible to mathematical treatment (1). Consequently, the mechanism of turbulent flow and of its transport processes must be considered in close conjunction with the available results of experimental investigation (2,3).

Turbulent transport data are obtained at two levels or classifications. The first group are the flux and profile data such as coefficients of friction, drag, and heat transfer, mean velocity and temperature distributions, the diabatic constant in the wind profile for nonneutral stability, and the constant of Kolmogoroff's hypothesis for the inertial subrange. The second group contains the more detailed characteristics of the turbulence such as mean-square values of fluctuating velocity components, temperature, and their derivatives, and the various correlations and cross correlations. Measurements of this second class of properties are practically nonexistent except for the case of channel flow with no temperature gradients (4).

Homogeneous isotropic turbulence has received most of the theoretical and experimental attention devoted to the subject of turbulent flows. The flows encountered in most problems of practical interest are seldom either homogeneous or isotropic and often have the added complication of a velocity gradient, together with gradients of density, temperature, etc. These gradients in turn bring about profound changes in the structure of the turbulence (5). In nonisotropic flow, time mean values



of shear stresses are present and serve to generate turbulent motions (6). Thus in contrast with isotropic turbulence, where turbulent motions have a decaying character owing to viscous dissipation, in nonisotropic flow there is production as well as dissipation of turbulence. Theoretical work in this area is being done (2,7,8), however, the need for experimentation is very real.

The investigation of various types of turbulent flow has received considerable impetus during the last decade because of the importance of the pressure fluctuations associated with these flows. The turbulent boundary layer and turbulent jet are of particular interest at the present. The use of correlation techniques for examining the turbulent velocity and pressure fluctuations must be made with some care in the presence of a mean shear and an appreciable turbulence intensity. This is due to the fact that turbulence is being continually created and modified by the shear stresses. Thus Taylor's hypothesis that turbulence may be regarded as a frozen pattern of eddies being swept past the observer is no longer strictly valid (9).

An increasingly wider variety of flow problems and applications of turbulent flow is appearing. The role of turbulence in coagulation of aerosols and colloids (10) has been studied for some time. Recently there has been an interest in turbulent fluid amplifiers for logic devices (11). Also turbulence is now believed to play a role in star formation in elliptical galaxies (12). The turbulent flow characteristics of non-Newtonian and viscoelastic fluids has been studied (13,14). It does appear, however, that viscoelastic fluids act to suppress turbulence and reduce the drag coefficients (14).

In the field of magnetohydrodynamics nearly all the properties of turbulence are beginning to receive attention. Drag and wake structure (15), heat transfer (16), electron diffusion (17), turbulent boundary layers (18), and the correlation coefficients and energy spectrum (19,20) have been considered. However, it will probably be a few more years before a substantial amount of good experimental data are available.

New methods and ways of applying turbulent transport data appear quite regularly and frequently. They range from analytic extensions of classical methods (21) for boundary layers and series expansions for heat transfer (22) and turbulence correlations (23) to numerical computer programs for the calculation of lift due to wind gusts (24) and the behavior of compressible boundary layers with variable temperature (25,26).

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## B. Confined Turbulent Flow Fields

When the structure of the turbulent flow field is directly influenced by the presence of a solid boundary it is said to be confined or wall turbulence. Such flow is nonisotropic and depends upon the nature and configuration of the boundary.

In general confined turbulent flows fall into two groups: one in which the flow is around or over a body, boundary layer flow; and, the other where the flow is within a space bounded by walls. The principal difference between the two groups is that, in the first, the region where turbulence occurs increases along the body in the downstream direction, whereas, in the second the turbulence remains constricted to the space between the walls. Most of the experimental data on these types of flows is for two-dimensional flow with zero downstream pressure gradient along a flat plate in the first case, and for flow through round tubes and between straight two dimensional channels in the second case.

### B-1 Ducted and Conduit Flow

In the entrance region of a conduit a boundary layer forms and grows in thickness in the downstream direction. The flow is said to be fully developed beyond a point downstream where the boundary layers from adjacent walls have merged (1). The conditions at the entrance of the conduit greatly influence the length required for the fully developed velocity profile to form (2,3).

For turbulent flow the distance required for the local friction factor to equal the fully developed friction factor is considerably less than that required for the development of the velocity profile (4,5). With disturbance free entry of the fluid into the pipe, transition to turbulence may occur if  $u x / \nu > 10^5$  (2). This is in general agreement with

the corresponding value for the boundary layer (see next section). When the entering flow is already turbulent, Latzko (3) calculated the distance using the 1/7 power law for the velocity to be

$$x/D = 0.693 \text{ Re}^{1/4}$$

This formula generally gives somewhat shorter values than are observed experimentally (2).

In addition to the entrance transition to fully developed flow it is necessary to mention the transition from laminar to turbulent flow that takes place as Reynolds number is increased. A considerable amount of both theoretical and experimental effort has been made on this problem (1,3). The transition from laminar to turbulent flow is accompanied by a significant change in the resistance and the velocity profile. It is generally found that the transition occurs at a Reynolds number of 2300 with full turbulence be achieved at Reynolds numbers greater than about 5,000. However, experimenters have succeeded in maintaining laminar flow at Reynolds numbers up to 40,000 (3).

In pipe flow there is no interaction with a turbulence free stream and no intermittency of turbulence as is found in boundary layer flow (2). Another dissimilarity is found in the fact that in fully developed pipe flow the conditions are independent of axial position, there is no mean radial velocity and the pressure is uniform across the pipe. Also the shear-stress and pressure forces are in equilibrium so that the shear stress varies linearly with radius.

The early attempts at correlation of the mean velocity profile for turbulent flow in pipes employed simple power dependency on distance from the wall. The 1/7 power being quite commonly used. Using his mixing length concept Prandtl developed the logarithmic velocity distribution. The improvements

in the universal velocity distribution by Deissler (6,7,8) take the form

$$u^+ = 3.8 + 2.78 \ln y +$$

for  $y^+ > 26$  and

$$y^+ = \frac{(1/\sqrt{2\pi}) \int_0^{0.109u^+} e^{-(0.109u^+)^2/2} d(0.109u^+)}{0.109 (1/\sqrt{2\pi}) e^{-(0.109u^+)^2/2}}$$

for  $y^+$  between 0 and 26, where

$$u^+ = u/u^*,$$

$$y^+ = yu^*/\nu, \text{ and}$$

$$u^+ = \sqrt{\tau_w g_c / \rho} = v \sqrt{f/2} \text{ for pipe flow.}$$

The data are in good agreement with these curves. Perhaps the only major objection is the fact that at the axis of the tube the logarithmic expression does not give a zero velocity gradient.

These equations also apply to open channel flow and Deissler (6) has shown that for adiabatic flow the effects of compressibility on the velocity distribution is small up to sonic velocity. Modifications have been proposed for the velocity distribution with heat transfer (6) and for rough pipes (9). Also in conjunction with the velocity profiles a number of correlations are available for friction factors in both smooth and rough pipes and in the presence of heat transfer (1).

Perhaps the most complete measurements of the structure of turbulence in pipe flow are those made by Laufer (10). He used a hot-wire anemometer to measure velocity profiles, the RMS of the turbulent velocity fluctuations  $\sqrt{u'^2}$ ,  $\sqrt{v'^2}$ , and  $\sqrt{w'^2}$ , shear stresses expressed as  $u'v'/u^{*2}$ , and turbulent energy spectra for air at Reynolds numbers of 50,000 and 500,000.

The work of Laufer (10), Reichardt (11), Townsend (12) and others (13,14,15,16,17) provides a picture of the structure

of turbulent pipe flow (2). To a certain extent the picture also applies to turbulent boundary layers, since there is a great similarity between pipe flow and boundary layer flow, particularly in the vicinity of the wall. The turbulent kinetic energy shows a sharp maximum close to the wall,  $y^+ \approx 12$ , which is the region of maximum production and dissipation. In the central portion of the pipe the turbulent energy balance is furnished by the production and dissipation so that the turbulence is practically in energy equilibrium. There is a flux of kinetic energy by turbulent diffusion toward the center of the pipe, causing a loss of energy in the wall region. At the same time the wall region experiences a gain in energy by turbulent diffusion of pressure energy from the core toward the wall.

The production of turbulent kinetic energy by the shearing action of the mean flows occurs mainly in favor of the  $u_x'$  component. Most of this energy is immediately dissipated, but a part is transferred to more wall-remote regions. Townsend (12) offers the explanation that turbulence near the wall consists partly of small attached eddies with a fixed orientation and partly of free eddies which diffuse by turbulence to the inner section. These free eddies transport their kinetic energy as they decay.

The energy spectrum of the axial component indicates not only that there is strong interaction with the mean motion, but also that the wave number range increases toward the higher values as the wall is approached. The large eddies elongated in the axial direction occur with a diameter roughly equal to one-quarter of the pipe diameter and with their main velocity in the axial direction. Small eddies are superimposed on these large eddies with the smaller eddies being near the wall.



There are relatively few investigations of the distribution of scalar quantities such as heat and chemical species (2). In particular there have been few measurements of the distribution of temperature fluctuations or their correlations with velocity fluctuations. Insight into the mechanism of the turbulent transport of a scalar quantity is thus based upon assumed analogy between the transport of a scalar quantity and transport of momentum. The usual practice is to seek correlations of transfer coefficients and where the scalar profile in available eddy diffusivities are determined from the analogy relations.

Azer and Chao (18) surveyed the eddy diffusivity data for heat transfer prior to 1960 and using a modified mixing-length model proposed the correlations.

$$\frac{E_x}{E_y} = \left\{ \frac{1 + 135 N_{Re} e^{-0.45 - (Y/R)^{0.25}}}{1 + 57 N_{Re} N_{Pr} e^{-0.46 - 0.58 - (Y/R)^{0.25}}} \right\}$$

for fluids of  $N_{Pr}$  ranging from 0.6 to 15, and

$$\frac{E_x}{E_y} = \left\{ \frac{1 + 135 N_{Re} e^{-0.45 - (Y/R)^{0.25}}}{1 + 380 (N_{Re} N_{Pr}) e^{-0.58 - (Y/R)^{0.25}}} \right\}$$

for liquid metals. The authors state that these equations fit the data with a maximum deviation of less than 14%. New data are becoming available and it may be necessary to make adjustments in the correlation (19,20,21,22,23,24,25). In addition mass transfer data on eddy diffusivity do not agree with the analogous form of Azer and Chao's equation (26,27,28,29). Alternate models (30,31) have been proposed based on the suggestions of Townsend mentioned above (12).

Data on transport under extreme conditions are also becoming available. Eddy diffusivities for forced convection to hydrogen and helium in the supercritical region have been reported (32,33). Heat, mass and momentum transfer in frosted pipes (34) and for gas-solid systems at high temperatures is also under investigation (35,36,37).

Interest in injection of a second stream into a supersonic stream has increased dramatically in the last several years (38,39). This is due primarily to the intensive work on Thrust vector control, fuel injection and reaction control. A number of different geometries and flow arrangements have been used (39,40,41,42), with the principle difference being injection in line or normal to the main stream. Eddy diffusivities for the spreading of the jets have been calculated from the data with proper allowance for total temperature and density variations (39,41). The mixing with combustion is now beginning to receive attention (43,44).

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## B-2 Turbulent Boundary Layers

When fluid flows past a body the velocity decreases from the free stream value to zero at the surface. This decrease in velocity takes place in a small region near the surface known as the boundary layer. At the front stagnation or leading edge the boundary layer is of zero thickness and becomes thicker as the flow progresses along the surface. For sharp and streamline leading edges flow in the layer is laminar at the front and becomes turbulent as the flow progresses along the surface.

In 1904 Prandtl suggested that in case of fluids of small viscosity the field of flow can be divided for the purposes of mathematical analysis, into two regions: the thin boundary layer near the wall, in which friction must be taken into account; and, the region outside the boundary layer, where the forces due to friction are small and may be neglected (1). Such a division of the field of flow brings about a considerable simplification of the mathematical theory of the motion of fluids of low viscosity.

The definition of the boundary layer thickness is to a certain extent arbitrary because transition from the velocity in the boundary to that outside it takes place asymptotically. From a practical point of view the convention has been adopted of defining the boundary layer thickness as that distance from the wall where the velocity differs by one per cent from the external velocity.

When fluid flows past a two-dimensional body, such as a cylinder, the fluid is accelerated as it passes over the forward portion of the body and is then decelerated after it passes the thickest part of the body. This effect causes separation of the boundary layer from the curved surface. Pressure increases over the forward portion of the body and decreases

behind the body. The reverse motion accompanying boundary layer separation and pressure decrease gives rise to a vortex. For values of the Reynolds number between 60 and 5000 it is possible to discern a regular pattern of vortices which is known as a Karman vortex street (1). At lower Reynolds numbers the wake is laminar and at higher Reynolds numbers there is complete turbulent mixing. Wake flows and other separated flows are considered in subsequent sections.

Some types of entrance flows were discussed in the previous section. The distinction of these entrance flows from other boundary layer flows was made because they share the common characteristic of merging into fully developed flow at some downstream position in a duct or pipe (2).

Transition from a laminar to a turbulent boundary layer may occur if the Reynolds number exceeds a critical value. Schubauer and Skramstad (3) determined the critical value of the Reynolds number,  $UX/\nu$ , for the onset of turbulence in the laminar boundary layer of a flat plate for different turbulence conditions of the free stream. When the relative intensity of turbulence in the free stream is below 0.1% it does not influence the transition which occurs at a Reynolds number of  $3 \times 10^6$ . At a relative intensity of 0.5% transition occurs at a  $10^6$  value of the Reynolds number. For higher levels of free stream turbulence (4,5) the transition is near  $5 \times 10^5$ . Recent studies of transition deal with hypersonic (6,7) and three dimensional flows (8).

A number of measurements on turbulent intensities in a boundary layer have been reported, but those on other turbulence quantities, such as shear stress, spatial correlations, dissipation, etc. are much scarcer (9). Experiments show that the intensities of the turbulence velocity components differ

appreciably within the boundary layer (10,11,12,13). In the wall region the shear stress is practically constant (12) and further out in the boundary layer von Karman's similarity between shear stress and kinetic energy distributions appears valid.

The distribution of the eddy viscosity has been obtained from the shear stress and mean velocity distributions (9,14). Near the wall  $\epsilon_y / u^* \delta$  appears to vary as  $y + 2$ . With increasing distance from the wall the eddy viscosity shows a linear variation. A maximum is reached roughly at  $y/\delta = 0.3$  after which the eddy viscosity decreases to the free stream value.

Current investigations of boundary layer flow include the structure (15) and influence of pressure fields (16,17,18). Also the influence of Mach number and other factors for high Reynolds numbers and compressible flow is being studied (19,20). What little information exists on compressible boundary layers is for the most part based on the transposition of concepts from incompressible flow theory (1,21).

As fluid flows past a solid body which is at a different temperature, a thermal boundary layer as well as the hydrodynamic layer forms. In the thermal boundary layer the fluid temperature at the solid surface equals that of the surface. The thickness of the thermal boundary layer may be different from that of the hydrodynamic boundary layer, it starts at one point where the heat transfer begins, which may or may not be the leading edge. For a Prandtl number of unity, both boundary layers grow at the same rate (22). For Prandtl numbers less than one, the thermal boundary layer grows more rapidly than the velocity layer. On the other hand, for Prandtl numbers greater than unity, the thermal layer grows slower than the



velocity layer. In a like manner mass transfer boundary layers have been observed (23) and the interpretation is identical.

Analysis of thermal and mass transfer boundary layers requires a knowledge of the velocity. Thus the equations of change are coupled and must be considered together. The major portion of the early work with thermal boundary layers was done by Pohlhausen (24). Both the direct approach to solutions and the integral approach is used (2,25,26).

Current investigations of thermal boundary layers deal with free stream (27), discontinuities (28), protubances (29,30) and surface motion (31). Hypersonic flow where viscous heating is important (32) and boundary layers with catalytic surfaces are receiving attention (33).

Mass transfer boundary layer work includes transpiration (34,35) and ablation (36,37). The effect of free stream turbulence (38) and mass transfer in the entry region (39) have been studied and the data interpreted in terms of eddy diffusivities.

Boundary layer flow with chemical reaction has begun to receive increased experimental attention. Combustion in turbulent boundary layers and associated transport properties have been reported (40,41,42). Also work on dissociation of gases is beginning to appear (43).

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### D-3 Flow in Special Geometries

Special geometries represents all geometries not included under the previous sections dealing with confined turbulent flow. This represents flows of very diverse nature, particularly as exhibited by the chemical process industries which employ a broad array of fluid handling equipment of differing geometries. It also includes some of the studies of special designs in aero-space systems that can not be conveniently located in other sections.

Experimental studies of fully developed turbulence properties and correlations in rectangular (1) and triangular (2) channels have been made. The special flow patterns characteristic of these cross sections exert some influence on the nature of the turbulent fields. Similarly there is some information on flow in curved channels (3). Townsend (4) has analyzed the response of a turbulent boundary layer to abrupt changes in surface conditions.

Investigations of flow in porous media have been conducted by petroleum scientists for some time and more recently by space scientists interested in cooling re-entry vehicles (5,6,7,8). Generally the flow is laminar through a random geometry, however, the mixing and near surface flow lends itself to turbulent analysis with the porous transpiration serving as a source of fluid.

Turbulent transport data for rotating discs (9,10) and concentric spheres (11) are available. Flow separation and heat transfer (12,13) in sharp corners has been reported. Also boundary layers and cellular patterns in transverse rectangular cavities (14), vertical slots (15) and behind a blunt trailing edge fitted with splitter plates (16) have been investigated.

Data on natural convection and stratification in vessels (17) is available. Heat transfer coefficients for turbulent film condensation have been correlated using the Deissler and von Karman representations of the eddy viscosity and a constant value for the turbulent Prandtl number (18). Heat and mass transfer in packed beds has been correlated using an eddy model for the turbulent contribution to molecular transport coefficients (19,20). Frequency response analysis of multipass shell and tube heat exchangers employs eddy diffusion models (21).

Rosenweig (22) studied turbulent mixing of miscible systems in a continuous flow stirred vessel. Watzen and Hubbard (23) analyzed the dynamic behavior of a pulsed-plate extraction column using a model using an axial eddy diffusion model for the pulsation back mixing. Eddy diffusivities for both liquid phases in an extraction column packed with rings or spheres have been reported (24). Systems used were kerosene in water, water in kerosene and water in mineral oil. The eddy diffusivities varied with phase velocities and the wetting ability of the discontinuous phase. Similar work with rotating disc contactors and other equipment have resulted in eddy correlations (25,26).

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### C. Free Flows

Heat, mass and momentum transfer in jets and streams issuing into large bodies, meteorological and oceanographic flows represent types of transport in essentially free or unconfined flows. These flows are distinguished by the absence of direct constraints on transport processes imposed by solid boundaries.

#### C-1 Free Jet and Wake Flow

Though jet and wake flow represent two types of unconfined flow much of their analysis conforms with that for boundary layer flow. This is so because the region in space for which a solution is sought frequently does not extend far in a transverse direction compared with the main direction of flow, and the transverse gradients are large. Quite often jets may be considered as point or line sources for purposes of analysis (1). The general problem of the round jet issuing in parallel flow has been treated by Squire and Truncer (2). Pai (3) has considered the problem using a value of the eddy viscosity that is constant across the jet and a function of downstream position only.

Jet flow occurs when a fluid is discharged as a confined flow, for example, from a pipe or through an orifice into another fluid which may itself be moving or at rest. The discontinuity in the velocity at the jet surface is unstable and gives rise to a zone of mixing downstream. Surrounding fluid becomes entrained in the jet stream. Consequently the mass flow rate increases in the main stream, the jet spreads out and its velocity decreases (4). Some aspects of plumes and jets entering the atmosphere are discussed in the subsequent section.

A wake is formed behind a solid body which is moving through a fluid at rest, or behind a solid body which is in

a stream of moving fluid. The velocities of a wake are smaller than those in the main stream and the losses in the velocity in the wake amount to a loss of momentum which is due to the drag on the body (4). When the flow in the boundary layer adjacent to a solid surface is reversed the boundary layer separates from the surface. Vortices form downstream from the separation. This region of strongly decelerated flow spreads as the downstream distance increases and the velocity approaches that outside the wake. The momentum lost due to boundary layer separation and wake formation is usually called form drag.

Measurements of mean velocity, shear stress, intensity and structure of turbulent flow in jets and wakes have been made (1,4). Some of the conclusions that have been drawn are: the lateral spread of a turbulent region with respect to the main flow direction is relatively slow; the turbulent region is separated from the nonturbulent region by an irregularly distorted boundary surface; the turbulent flow has an intermittent character, in particular toward the average boundary of the turbulent region; many properties, such as turbulent intensities and dissipation, show great uniformity across the main body of the turbulent flow, decreasing essentially to zero only in the intermittent boundary region; and, this uniformity is increased over a larger region if the distribution function of the quantity considered is corrected for the intermittency factor.

The structure of self-perseving jets (5,6) has been receiving increased attention. Self-perservation of the over-all structure as shown by the velocity and increase in width occurs at appreciable distances downstream as indicated by the extensive work of Townsend (7) on wake flow behind cylinders. Turbulent mixing of coaxial jets (8), supersonic

and subsonic jet (9,10) and axisymmetric swirling jets (11) are actively being studied (12). Some discussion of confined jets and wakes was presented in the previous section.

Heat transfer (13,14) and mass transfer (15,16,17,18) properties of free jets have been studied for both gas and liquid systems. The turbulent Schmidt numbers agree with those observed for pipe flow (19,20). Studies of the break up of liquid jets (21) and liquid fuel injection using eddy diffusivity interpretations of the data have been made (22). In addition data have been reported on supersonically convected turbulent eddies and the nature of turbulence in the noise producing region of circular jets associated with rocket and aircraft engines (23,24). And some information on flow, heat transfer and turbulence in Ranque-Hilsch (vortex) tubes is available (25).

Townsend's experimental studies of wake flow brought together the momentum and vorticity transport approaches and provides the basis for subsequent work (1,7). Investigations of wakes in hypersonic flow fields (26,27), the structure of turbulence and transition (28,29,30) are available. Distorted wakes (31,32,33) and wakes with chemical reaction (34) are being studied.

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C-2 Meteorological and Oceanographic Flows.

Much of the interest in turbulence for the meteorologist lies in its transport properties, notably in the transport of sensible heat, momentum, and water vapor or heat in latent form (1,2). In the lower atmosphere the small scale turbulent flux of these quantities in the vertical generally far exceeds that in the horizontal (3,4,5).

In the preface of his book (6) on the hydrodynamics of Oceans and Atmospheres, C. H. Eckart of the Scripps Institute states: "The following pages will contain no account of the effects of viscosity and turbulence, which modern meteorology and oceanography invoke so freely to explain many phenomena. The reason is that, in geophysics, there are two Reynolds numbers. The first is the ordinary one:

$$R = VL/\nu$$

V being a typical velocity, L a typical length and  $\nu$  the kinematic viscosity. The other is:

$$R_g = NL^2/\nu$$

N being the Vaisala-Brunt frequency (which may be thought of as summarizing the small scale dynamics of an adiabatic expansion). In laboratory experiments,  $R_g \ll R$ , and the latter will dominate the phenomena. For large, or moderately large scale phenomena in geophysical fluids,  $R_g \gg R$ , and the former will dominate. Consequently it is most improbable that the results of laboratory investigators of turbulence will have much relevance to turbulence in the air or oceans. In particular, the theory of isotropic turbulence will have little application to those fluctuations of the wind that disturb the even flight of an aircraft."

Turbulent dissipation effects are negligible in the analysis of the large scale motion of oceans or the atmosphere. In

smaller scale problems two kinds of dissipation can be distinguished (6). The first is sometimes called the spreading of wave packets and occurs whenever the velocity of simple waves in a medium is a function of frequency. This phenomena is not generally classified as turbulence. The second results from the coupling between eigen solutions, induced by (1) the non-linearity of the basic equations and (2) the small scale topography of the boundaries. Our attention then is directed to the latter phenomena.

In research on the turbulent structure of the lower atmosphere the major complication not found in laboratory studies of fully developed channel flow are associated with the absence of upper and lateral boundaries, and with broader ranges of space and time scales of turbulent motion (7). On the other hand, the all important semblance between the two fields of study is the appearance of the same indeterminate quantities in the equations describing the mean flow, mean temperature, turbulent kinentic energy, etc. in both cases.

Near the surface of the earth it is generally found that the three fluxes are effectively constant with heights from the top of the air layer, where molecular and radiative effects are significant, up to a height of several meters, or even during the main hours of insolation, to 100 meters or so (1). Values of the eddy diffusivity around 2500 cm<sup>2</sup>/sec have been found to be typical at 1.5 meters over grasslands of about 4 m/sec and slight to moderate instability.

Swinbank (1) and Mistry (9) have correlated the turbulent Prandtl number with layer stability in terms of the Richardson number,  $g(\frac{dT}{dz} + \frac{\rho}{T}) / T(\frac{du}{dz})^2$ , which represents the ratio of the rate of consumption of energy by the buoyancy forces to the rate of its production by wind shear. For Richardson

numbers of zero, the turbulent Prandtl number is about 1.4 and for a Richardson number of -0.3 it is about 0.55. Also within experimental error the values of the turbulent Schmidt number for water diffusing through air has been found to be unity.

The form of the wind profile and its relation to the shearing stress has been the source of a number of investigations (1,10). Aerodynamically smooth flow is said to exist when the profile and shear stress are related to the kinematic viscosity. At heights above  $Z = 30 \nu / u^*$  the profile is given by

$$u/u^* = (1/k) \ln (u^*Z / \nu) + \text{constant}$$

for the mean velocity with the von Karman constant  $k$  being about 0.4. Below this level the shearing stress is partly turbulent as the surface is approached and turbulence ceases at the surface.

With increase in wind speed over a given surface or as the surface becomes increasingly rough at constant wind speed, a state is reached at which the purely viscous stress at the surface is outweighed by the effect of pressure forces associated with eddying wakes from the roughness of surface elements. The flow is then aerodynamically rough and the viscosity ceases to influence the profile, which is found to depend on a length which is characteristic of the surface roughness.

$$u/u^* = (1/k) \ln (Z/Z_0)$$

The roughness length,  $Z_0$ , is generally an order of magnitude smaller than the actual height of the roughness elements.

Aerodynamically rough flow is found to occur when  $u^*Z_0/\nu > 2.5$ . The two profiles are similar in form when  $\nu$  is replaced by  $u^*Z_0$ , the macroviscosity (11).

At any significant wind speed the flow will be aerodynamically rough over any vegetated surface, though smooth flow may exist



over snowfields and open water in the lighter winds. Whether the flow is smooth or rough the eddy diffusivity is given by

$$E_d = k \overline{w'z'} = k u^* z$$

Also it is convenient at times to introduce a drag coefficient

$$C_D = \tau / \rho u^2 = (u^*/u)^2$$

Values of  $C_D$  range from 0.001 for snow to 0.010 for grass.

Under adiabatic condition at sea the roughness will depend primarily on the wind speed. To the extent that this dependence is complete, the wind profile will contain only a single parameter, the wind speed at one height, and this conclusion has been somewhat supported by experiment (12). However, more data are needed to resolve the dependency of the drag coefficient or wind speed and the transition between smooth and rough flows (13).

When the air is being heated or cooled from below, the effects of thermal stratification and bouyancy must also be considered so that at least one more basic parameter is required (14). Batchelor (15) has shown that the Richardson number characterizes full dynamic similarity for these conditions and it has been used as the additional parameter by Monin and Obukhov (1).

The geostrophic wind involves deeper layers and is governed by the way in which momentum is absorbed as a function of height. Townsend (16) has shown how internal waves may be excited by a turbulent boundary layer. Thus in a barotropic fluid, such as the tropopause, a fluctuating potential flow may be induced by the turbulent boundary layer on the earth's surface which would provide energy for the onset of wave motions at higher elevations.

In unaccelerated horizontal motion the velocity vector  $\vec{V}$

is given by the equation

$$\lambda (\vec{k} \times (\vec{V} - \vec{J}) = \frac{\partial}{\partial Z} \left( \epsilon \epsilon, \frac{\partial \vec{V}}{\partial Z} \right)$$

where  $\lambda$  is the Coriolis parameter,  $\vec{J}$  the geostrophic wind, and  $\vec{k}$  a unit vertical vector (1). Both theoretical and experimental approaches have been taken at this point. In the first case some model for  $\epsilon_v$  as a function of  $Z$  is assumed and the above equation solved for  $\vec{V}$  as a function of  $Z$ . This gives a relationship between the wind and the isobars which is then checked against observation. At the present time it seems that the most realistic model is that of Rossby and Montgomery (1) who assume a surface-layer law of the form

$$\epsilon_v = k u_* Z$$

to hold up to some height and thereafter a steady decrease of  $\epsilon_v$  to a value of  $(\epsilon_v) = 50/\text{cm sec}$  which would represent the residual turbulence of the free atmosphere. The second approach is to adopt a position of ignorance about the transfer mechanism and use the height integral of the equation to evaluate data. The problem is complicated by the range of variation which the thermal wind regime and the daily incidence of convection and other symptoms of thermal stratification can impose (17).

In the study of the spectrum of turbulence and the structure of free convection in the atmosphere it seems to be a characteristic of the data that the small eddies contribute less to the heat and momentum fluxes than to the energy, and the largest eddies more to the heat flux than to the others. The marked increase in eddy size with height is also characteristic (18, 19, 20, 21).

Cramer et. al. (22) have measured the integral scale of turbulence and turbulent energy dissipation rates at 16 and 40 meter elevation above a smooth land surface. At higher elevations, from calculations of atmosphere energy, Haines and

Kruger (23) report that the atmospheric eddy generation fluctuates from about  $-0.1$  to  $-1.4$  watts/m<sup>2</sup> between summer and winter.

In the jet stream eddies typically have horizontal dimensions of 20 to 600 miles; and exchange coefficients of the order of  $10^9$  cm<sup>2</sup>/sec, i. e.,  $10^9$  stokes, have been evaluated (24).

Winds and turbulences are being studied by chemical release (25,26). Winds as high as 400 ft/sec are frequently observed and wind shear of about 0.02 ft/sec extending 10,000 to 20,000 ft. in altitude are common. Considerable turbulence is observed in the altitude region 200,000 to 320,000 ft. (27). Above 370,000 ft. the atmosphere appears quite stable. This is probably due to the eddy Reynolds numbers becoming quite small and to the large positive temperature gradients resulting in Richardson numbers becoming large. At 300,000 ft. the diameters of large scale eddies are as large as 6000 ft. and eddy diameters as small as 10 ft. have been observed.

The statistical properties of atmospheric pressure variation were evaluated by Richie and Hixan (38), up to pressures of 500 dyne/cm<sup>2</sup> and frequencies of 0.033 to 50 cycles/sec. Several numerical experiments have been made to provide connections between the eddy transport properties and other parameters associated with atmospheric turbulence (29,30, 31,32). Blackadar has correlated the eddy viscosity in a neutral atmosphere (33) and Wu (34) has used an eddy mechanism for heat transfer data in the lower atmosphere.

Large free burning fires and other sources of heat cause plumes or thermal jets of hot gas and air that rise into the atmosphere. These local updrafts can carry particles and heat over large distances. Fire plume models have been proposed (35) and data for different kinds of plumes are available (36,37). The analysis of plumes is identical to that used for free turbulent jets. Data are available on the starting

portion of plumes (38) and for hot gas plumes in horizontal winds (39).

The amount and extent of turbulent transfer data for oceanographic flows appears to be at least of an order of magnitude smaller than that for meteorological flows (2,40). Horizontal mixing in the sea due to shearing currents (44) and in small scale experiments (41,42,43) have been reported. Some data are available on the turbulent spectrum in tidal streams and channels (45,46).

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